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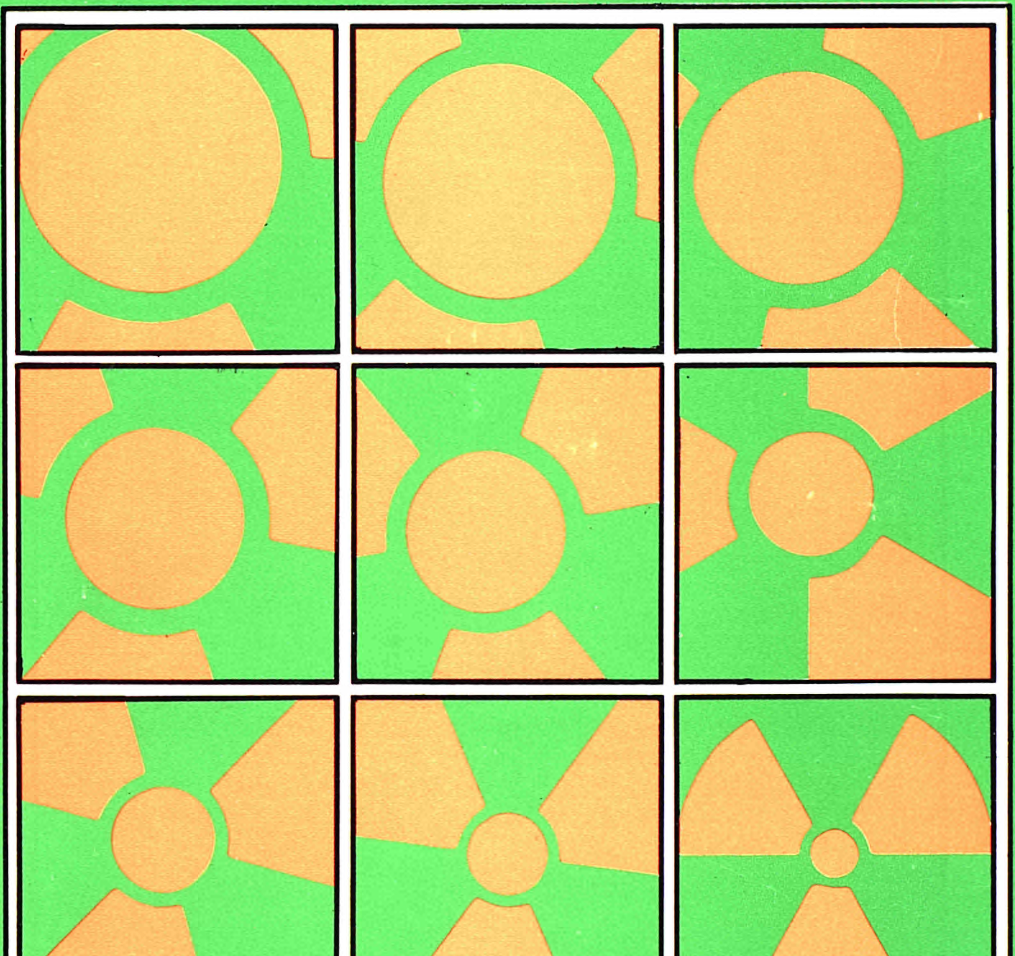
nuclear science and technology

Field studies about radionuclide migration, natural analogues and faults in clays



Report

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Field studies about radionuclide migration, natural analogues and faults in clays

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Final report for CEC contracts
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FINAL REPORT FOR CEC CONTRACT

FI - 1W - 0064(UK)

July 1986 to June 1989

In situ determination of the effects of organics on the mobility of radionuclides in controlled conditions of groundwater flow.

G. M. Williams

1. RESEARCH OBJECTIVES

(a) To undertake a literature review to collate the available information on the character of organics arising from the burial of low level waste in shallow ground, and to review the present knowledge of the effect of organics on radionuclide mobility in the geosphere.

(b) To identify, and characterise, a field experimental site in a confined glacial sand aquifer suitable for *in situ* tracer tests. This will be within an area of the British Nuclear Fuels premises at Drigg in Cumbria.

(c) To characterise the physical and chemical properties of core material and natural groundwater, and to undertake laboratory sorption experiments to provide data to enable predictions to be made of the outcome of a field tracer test with selected radionuclide species. In particular the effect of natural organics (humic acids) in the groundwater on radionuclide mobility will be studied.

(d) To use available geochemical models to determine the speciation and mobility of radionuclides in the presence of organic compounds identified in (a) above, so that their behaviour in a tracer test can be predicted.

(e) To undertake tracer tests with selected organics in addition to those naturally present in order to determine their effect on radionuclide mobility.

(f) The CEC also requested BGS to participate in the COCO club, set up by the CEC, comprising researchers studying the behaviour of complexes and colloids in groundwater.

2 ORGANISATION OF THE RESEARCH PROGRAMME

The work has been carried out under a joint contract from the UK Department of the Environment and the Commission of the European Communities (CEC) . The work has been undertaken by BGS in collaboration with Loughborough University of Technology (LUT), and the University of Wales College of Cardiff (UWCC). Loughborough University has been involved in direct speciation determinations of radionuclide organic complexes, while UWCC has undertaken speciation modelling. The CEC stipulated that formal liaison over modelling the field experiment should be established with Delft Geotechnics in the Netherlands, with no financial agreement between BGS and Delft Geotechnics.

3 SUMMARY OF RESULTS.

3.1 LITERATURE REVIEW

A literature review of the effects of organics on radionuclide mobility has been produced (Ross and West, 1985). Published literature from this review was been sent to Carlsen (Riso laboratory) who has compiled a comprehensive report of the effects of organics on radionuclide mobility on behalf of the COCO group (Carlsen 1988). During the course of the contract it became apparent that there was also a need for a review of models to predict the interaction of cations with natural organic material and this has formed the major part of BGS activity (Falck, 1988).

3.2 CONSTRUCTION AND CHARACTERISATION OF A TRACER TEST ARRAY

3.2.1 Facilities at the *in situ* field laboratory

An array of boreholes has been constructed in a shallow confined glacial sand aquifer at the Drigg Storage Depot in Cumbria, after the exact location had been identified by a resistivity penetrometer survey (Jackson et al, 1989). The borehole array is enclosed in a prefabricated wooden hut with mains electricity connected, which can be dismantled for future drilling/hydraulic testing purposes. The field laboratory also comprises 2 portacabins; one forming an office/amenity facility, the other for radio-analysis instrumentation and for trace radio-active work. Mains electricity and water are available. The site is considered of long term value for future experiments beyond the current contract.

3.2.2 Drilling techniques and instrumentation

Special drilling and sampling methods were developed to construct the field tracer array with minimal disturbance of the aquifer (Wealthall et al, 1988). Groundwater is re-circulated between two boreholes 3.41 m apart and the tracer(s) are released in a third "on-line" borehole 0.94 m from the recharge well. The movement of radiotracers is monitored by removing groundwater from three multi-level sampler installations, using a vacuum extraction system, (Hitchman, 1988). Tracer migration is also monitored by means of three gamma probes on automatic ranging winches controlled by micro-computer, and connected to a multi-channel analyser (Williams, Roberts and Wealthall, 1989, Mooney and Roberts, 1989).

3.2.3 Lithology, mineralogy and physical properties

Cores recovered from the array have been logged and their mineralogy determined by X-ray diffraction, and evolved gas analysis. Particle size distributions and cation exchange capacities have been determined with depth. Cores have been frozen in liquid nitrogen so that relatively undisturbed sub-cores could be obtained parallel to the bedding for diffusion and permeameter experiments. Cores have also been treated with resin and prepared as thin sections for analysis by SEM and optical microscopy.

The sediment is composed principally of quartz, with subordinate amounts of orthoclase and plagioclase feldspars (Bloodworth and Inglethorpe, 1987). The clay content varies widely within the array but is constant in character being composed predominantly of mica, kaoloinite, and chlorite. Calcite and dolomite corellate with the clay fractions and do not exceed 5 % by weight. Organic matter lies between 0.1 - 2.5 %. Cation exchange capacity is low at around 1 - 4 meq/100g with up to 11.8 meq/100g in the clay rich fractions.

3.2.4 Groundwater chemistry

Groundwater within the array has been obtained from the multi-level samplers and other available boreholes, and analysed by ICP-OES and IC (Haigh et al, 1989). Samples of groundwater were also obtained during a pumpimg test and formed the basis for geochemical modelling. Samples were taken from the main flow path using in-situ multi-level sampling points. These samples enabled a profile of major species to be obtained. Groundwater from the array was of low ionic strength with major ions including calcium, magnesium, potassium, sodium, chloride, bicarbonate and sulphate. Redox potentials were determined using a platinum electrode and by analysis of redox couples. Differences in redox potential observed from these methods inferred that the system is not at redox equilibrium. The groundwater was found to be under-saturated with respect to calcite, gypsum and dolomite. The total organic carbon concentration was relatively constant in the sand horizon (6-12 mg/l) but significantly higher in the silt/clay horizons (20-70 mg/l).

A bulk groundwater sample (2 x 500 litres) has also been taken from the array for use in laboratory experiments. One of these bulk samples has been equilibrated with 50 Kg of glacial sand. Aliquots from both groundwaters were analysed periodically and the results compared to the groundwater collected from the sampling points in the array. Both bulk groundwaters exhibited a slightly lower concentration of major ions compared to multi-level sampling points within the array. The concentration of cations and bicarbonate was lower in the bulk groundwater containing sand, but chloride and sulphate were higher.

3.2.5 Geomicrobiology

The types, and to some extent, the numbers of different groups of micro-organisms present within the test zone of the aquifer have been determined (West et al, 1988). Analyses for total heterotrophs (including oligotrophs); fungi; sulphur, nitrogen and iron cycle microbes were performed using techniques designed to prevent contamination of samples in the field and in the laboratory. However, the numbers obtained do not necessarily represent the populations present in situ, and their activity was not measured. Microbes were found in all core and water samples, but numbers and types varied with depth and borehole. The most noticeable variation is the fluctuating, mostly low heterotrophic microbial populations in the silt/sand sequence found near the bottom of the aquifer test zone. Higher and more varied populations are found outside this zone. The variability in the distribution of fungi, sulphur cycle bacteria and iron oxidisers again seems to be related to the lithology. However, the absence of these forms were also noted within the aquifer. This could be related to Eh conditions or availability of nutrients required in particular by chemotrophic microbes such as sulphur oxidisers or iron oxidisers. Sulphide was present in very low amounts (0.006 mg/l) in every borehole sampled; iron oxides were present in varying amounts.

The study has given rise to a vast culture collection which can be used in future experiments. It is recommended that certain cultures are introduced into experiments where their influence on sorption of radionuclides can be studied. It would also be prudent to ascertain biomass *in situ* by simple calculations based on nutrient and energy supplies. Such models would indicate the effect of microbial activity on the degradation of any introduced organics within the array.

3.2.6 Colloid content

The concentration of natural colloids have been assessed in groundwater samples taken from the BGS experimental site at Drigg (Longworth et al, 1988). SEM analysis of filters yeided values of 10^8 - 10^{10} particles/litre. Mean mobilities of these particles determined by electrophoresis were in the range - (1-2) x 10^{-8} $m^2s^{-1}v^{-1}$. Uranium series disequilibrium measurements were used to determine the partition of natural actinides between particulate, colloid and solution phases. The activity of the colloid phase were found to be 0.1 - 0.5 % for uranium and 1 - 5 % for thorium.

3.2.7 Hydraulic characterisation

While constructing the array, profiles of hydraulic conductivity were determined at 75 mm intervals at 3 locations, giving a range of values between 1 x 10^{-6} and 1 x 10^{-5} m/s. A conventional pumping test gave K as 10^{-6} m/s and porosity (approx 0.35) has been determined from core material. Transient pumping tests have also been undertaken whilst rapidly monitoring multi-level samplers. This has provided values of hydraulic diffusivity (T/S) of approximately 150 m^2/s.

3.3 EXPERIMENTAL PROGRAMME TO DETERMINE RADIONUCLIDE/ ORGANIC/ SEDIMENT INTERACTIONS

3.3.1 Stability of anthropogenic organics

Acetate and EDTA have been encountered in leachates from intermediate and low level radioactive wastes (Ross and West, 1986) and have therefore been chosen for study in *in situ* experiments. Initailly a review was made on the degradation of these organics. Acetate ($CH_3.COO^-$) is involved in biological energy production and cell synthesis. It also plays an important part of the carbon geochemical cycle and in diagenetic processes. In continuous flow laboratory column experiments under aerobic and methanogenic conditions using acetate as a primary substrate plus other secondary substrates, influent concentration (1-100 mg/l) dropped by 95 to 99% after detention times of between 20 minutes to 2 days. Thus it appears that acetate will probably be short-lived in a subsurface environment. However, as acetate is such an important part of basic biological and geochemical processes, degradation rates can only be regarded as specific to a particular system. Degradation rates will vary with, for example, microbiological content, redox conditions and nutrient fluxes. Therefore it is prudent to view the above rates of acetate useage as indications only of what is likely to occur in the tracer test. Since the tracer experiment will probably be of relatively short duration any acetate added will probably make little impact on the system. However, any acetate remaining in the aquifer will be available for microbiological use possibly influencing future tracer experiments. This should be assessed by performing *in-situ* tests and monitoring acetate disappearance. Simple modelling calculations could also be performed.

EDTA is a multidentate chelating agent used as a means of decontamination/removal of radionuclides in various nuclear operations. As a result of its strong metal-binding properties EDTA is mobilising radionuclides in radioactive waste sites in the USA and thus poses a serious problem in waste containment. It is also extremely persistant in the environment being resistant to decomposition by radiation and thermally stable to 200 - 250°C. It is, however, sensitive to photolysis with by-products including glycine, IDA (iminodiacetic acid), possibly EDMA (ethylenediaminemonoacetic acid) and EDDA (ethylenediaminediacetic acid) which are more easily degraded than EDTA. The biodegradation of EDTA is slow, varying from 0.1% to 21% over 4 weeks in variously amended soils. Biodegradation can be slowed by the use of microbial inhibitors and the addition of sugars and amino acids and stimulated by the addition of sugar and ethylenediamine. In studies examining the influence of environmental factors on EDTA biodegradation in soils and sediments, biodegradation took place slowly under aerobic conditions and little took place under anaerobic conditions. EDTA intermediates, such as ethylenediaminetriacetic acid (EE3A), N,N-EDDA and EDMA, do not accumulate in soils and thus the rate-limiting step appears to be the initial degradative phase. However, despite the laboratory data, field studies do show that EDTA persists in the environment.

3.3.2 Batch tests

Batch experiments have been used to assess the effect of natural organics and EDTA, and acetate on the sorption of Cs, Sr, Co, Eu, Np, U and I (Haigh et al, 1988(a) and 1988(b)). Cobalt was chosen because it is relatively insensitive to redox changes, is known to form complexes with organics and could thus be used in technique development. The effects of natural organics were assessed by comparing experimental results using natural groundwater with those in which the organics had been removed from the groundwater by treatment with DEAE cellulose. Results show that the removal of natural organic material from the groundwater increased the distribution ratio of cobalt by a factor of about two. This suggests that the natural organic fraction removed by DEAE probably complexed with the cobalt to form an anionic species which was not sorbed. The effect of the residual organic material not removed by DEAE, is unknown.

The addition of both EDTA and acetate to the sand/groundwater system led to the removal of Ca, Mg, Sr and Ba from solution. Both organic compounds had the effect of reducing the pH and bicarbonate concentration probably due to the precipitation of the cations as carbonates. EDTA had the added effect of removing iron from solution. EDTA had a dramatic effect on the sorption of Co even at low concentrations reducing the R_d by up to 2 orders of magnitude. At concentrations greater than 5 mg/l, there was no further reduction in sorption. Experiments with radio-carbon labelled EDTA showed that EDTA was not sorbed, but formed a mobile complex with Co. Differences in distribution ratios were observed depending on whether the cobalt was added before or after EDTA. Within the same period of equilibrium, the R_d values were lower when EDTA was allowed to equilibrate with the sand before adding cobalt. The desorption of cobalt fom the mineral surface may be a rate limiting step such that equilibrium was not acheived within the 14 day period of these experiments. Acetate had no effect of cobalt sorption in any of the experiments undertaken. Implications of these results for possible tracer tests at Drigg include the need to assess the stability of the natural organic/cobalt complex and to consider if it could be separated for injection as a tracer. The stability of the natural organic complex(es) is important to assess if a tracer test can be carried out over a reasonable timescale i.e. before dissociation occurs. The Co-EDTA complex would be expected to behave as a conservative tracer in an *in situ* migration test. Again the stability of the complex needs to be assessed.

Strontium, caesium and iodine do not form strong complexes with organic ligands and their distribution ratios were not significantly affected by the addition of EDTA or acetate to the groundwater. The removal of natural organic material with DEAE cellulose also had no effect on the distribution ratio. Distribution ratios for neptunium were between 15 and 30 ml/g in oxic conditions at pH 8.3. The addition of high concentrations (3×10^{-3}M) of EDTA did not decrease the distribution ratio although speciation modelling had indicated that complexation should be nearly complete at EDTA concentrations of 3.5×10^{-3} M. Neptunium could conceivably be used in a field tracer test if automatic monitoring equipment were used but no suitable tracer is available.

The distribution ratio for uranium is pH dependent. It was about 260 ml/g at pH 6.0 and and 60 ml/g at pH 7.7. Speciation modelling showed that, as for neptunium, high concentrations ($\geq 10^{-3}$ M) of EDTA are needed before U-EDTA complexes are formed and complexation with the low concentrations of natural organic material in Drigg groundwater is, therefore, unlikely to occur (since the binding strength of humic acid is lower than that of EDTA complexes). These high distribution ratios and the fact that a suitable uranium isotope for long-term tests is not available, makes uranium unsuitable for field tests.

Europium was used as an analogue for the trivalent actinides. The ^{152}Eu used contained carrier and colloids were formed even at the minimum detectable concentrations of 3×10^{-9} M Eu. Distribution ratios were determined using solutions which had been filtered through 5000 MWt filters before and after spiking with europium. This procedure may have removed both true colloids and colloidal particles onto which europium had sorbed (pseudo-colloids). In spite of this, distribution ratios were high at >1000 ml/g. The addition of 3×10^{-6} M EDTA reduced the distribution ratio to 5 ml/g and increasing the EDTA concentration did not decrease it any further. It is postulated that some dissociation of the EDTA complex occurred during the equilibration period. Further work with trivalent lanthanides or actinides is indicated.

3.3.3 Direct speciation determinations

Radionuclide complexation with natural organic material in groundwater has been assessed by separating the "free" from "complexed" radionuclide species using gel chromatography (Warwick et al, 1988). A relatively non-invasive method has been used to separate complexed from free cobalt-60 in groundwater, using the weak cationic adsorption properties of Sephadex gel, and a mobile phase of natural groundwater. Results show the kinetics of Co complex formation in groundwater to be slow, and that the equilibrium position is affected by temperature, cobalt concentration and the ionic/organic strength of the groundwater. Treatment of the groundwater with DEAE cellulose to remove humic material, also removed the majority of organic species which absorb ultra violet at a wavelength of 254 nm, but 45% of the original total organic carbon remained, and the amount of complexed cobalt left in solution was only reduced to 76% of its former concentration. This suggests that the complexed Co species separated by the method described are a mixture of inorganic and organic compounds.

3.3.4 Diffusion experiments

A series of diffusion experiments using the through diffusion and infinite couple geometries have been conducted with ^{36}Cl, ^{125}I, ^{58}Co and ^{58}Co/(^{14}C) EDTA. Under sterile conditions iodine moved at the same rate as Cl. Under non-sterile conditions I was immobile and the influence of microbial activity is suspected (Higgo et al, 1989). The diffusion of cobalt in groundwater was compared with that of Co/EDTA in infinite couple experiments. In the former a two species diffusion profile was determined; in the latter only one species was present. The mathematics of diffusion with dissociation kinetics needs to be considered in interpreting experiments where two species are present.

3.3.5 Column experiments

Column experiments were used to simulate the migration of various organic/radionuclide complexes in the field tracer test and to identify the dominant sorption mechanisms (Higgo et al, 1989). Cobalt complexed with natural organic material (fulvic acid) in the groundwater, dissociated during migration through a column containing Drigg sand. Once dissociated, the free Co^{2+} is assumed to sorb strongly and rapidly onto the sand. The kinetics of dissociation were determined by sectioning the column and establishing the distribution of Co. A rate of 1.81×10^{-5} sec $^{-1}$ was inferred for the dissociation reaction. The Co/natural organic complex would therefore not be expected to migrate a significant distance if introduced into the field experiment.

Cobalt /EDTA complex (^{57}Co and ^{14}C EDTA) was passed through columns of Drigg sand at flow rates of 1.94×10^{-5}m/s and 2.78×10^{-5} m/s. Again the Co activity was attenuated but not to such an extent as with Co-FA. A complicated distribution of activity was observed along the column after passage of Co-EDTA suggesting that a number of Co species (possibly colloids) were present. Attempts have been made to simulate the distribution of Co in the column experiments assuming the presence of a number of species/ colloids. Results assuming the Co-EDTA to dissociate and form separate species each with a different retardation coefficient and dispersivity, have showed good agreement with the distribution of activity observed. However, additional work is required to confirm the mechanism for Co-EDTA dissociation.

3.4 GROUNDWATER FLOW, SOLUTE TRANSPORT AND GEOCHEMICAL MODELLING

3.4.1 Hydraulic modelling

The use of borehole pumping/recharge duplet, produces parallel flow lines in the vicinity of tracer release and monitoring wells. Consequently, 1-D and 2-D advection/dispersion models can be used to predict groundwater flow and solute migration rates (Sen and Williams, 1989). The test has been modelled initially in 1-D assuming that it is composed of a single homogeneous layer of thickness 1 m and porosity 0.35. Predicted travel times at a re-circulation rate of 2 l/min vary between 9 and 29 hours from nearest multi-level sampler to recovery (ie pumping) well. The flow velocity field has also been predicted by Delft Geotechnics using a multi-layered model based on field hydraulic profiles (Visser and Van Meurs, 1988).

3.4.2 Solute transport modelling

Solute transport modelling has been based on the results of laboratory column experiments and incorporates a relatively low longitudinal dispersivity appropriate to columns as determined in previous tracer tests (Williams et al 1985). Solute reactions that have been considered include simple reversible sorption to give a retardation factor, and dissociation of the mobile complex at a given rate proportional to the concentration which is similar to radioactive decay. Reactions involving sequential kinetically controlled stages are also being considered.

3.4.3 Geochemical modelling

Geochemical modelling using the PHREEQE code has been undertaken to determine the speciation chemistry of the Drigg groundwater and to identify the factors which affect the mobility of Co and Ni. The measured chemical composition of the groundwater has been used with an imposed range of redox and pH conditions, and in the presence of EDTA. (Falck et al, 1989).

Results suggest that the precipitation of calcium carbonate is unlikely to occur which precludes the co-precipitation of Sr or Cs. For the non-transition metals the free ion is the dominant aqueous phase species. This is not so for the transition elements eg for Mn the carbonate species is most abundant. Iron is sensitive to Eh/pH changes. $Fe(OH)_3{}^0$ dominates at Eh +227 mv to be replaced by Fe^{2+} at +117 mv with an order of magnitude increase in concentration. Co^{2+} is likely to be the dominant species under these conditions and would be sorbed strongly by the sediment. Ni, however, is redox insensitive, but a pH change from 6.0 to 8.0 is accompanied by a fall in Ni^{2+} composition from 71.3% to 3.6% with a concomitant increase in $NiCO_3{}^0$ from 19.6% to 95%. EDTA must be added at concentrations > 2.9 mg/l to maintain at least 90% complexation of Co if added at a concentration of 1.34×10^{-10} mol/l. At lower concentrations of EDTA, other trace metals would compete with Co for the ligand.

3.4.4 Modelling interactions between natural organics and cations

The techniques available for modelling the interaction between natural organic matter and metal cations/protons has been reviewed (Falck, 1988). These natural organic materials are macromolecules which exhibit a polyelectrolyte character owing to numerous dissociable functional groups which are attached to their carbon backbone or form integral parts of the structure. The polyelectrolyte character is thought to be responsible for their conformation, hydrogen bonding or bridging by metal cations between subunits being important mechanisms. Environmental parameters, like pH and ionic strength thus will have profound effects on the conformation of natural organic matter, the properties of which can change from being a flexible polymer to being a rigid gel.

Covalent and coordinate bonding between organic matter and cations may lead to complex formation. In cases where two or more ligands on the same organic molecule are available in suitable positions chelates may be formed. Electrostatic secondary forces resulting from the polyelectrolytic character may also be important. Approaches to model the interaction between cation and organic macromolecules can be divided into two groups: discrete ligand models and continuous distributions models. Discrete ligand models assume a known concentration of known ligands with known properties (like stability constants) and model the distribution of cations/protons between those and possibly inorganic ligands in very much the same way as do the well known speciation codes. Thus they are readily compatible with the latter. They only take into account true binding unspecific electrostatic interaction being neglected. Conformational changes due to changes in the physico-chemical environment are also neglected. In general these models do not consider the structure of the molecule(s). Random-structure models build a great number of molecular structures with functional groups attached to them obeying certain constraints (such as molecular weight, isomeric arrangements, branching of aliphatic chains etc.) and taking analytical results (elemental composition, aromaticity, functional group concentration etc.) into account. The resulting binding site distribution is analysed statistically and used as input into speciation model as in the case for other discrete ligand models. Again secondary forces are not considered.

Continuous distribution models are mainly employed to analyse experimental (titration) data and are distinguished by the transformation of the data set the analysis is performed on (raw titration curves, cumulative curves etc.). The aim is to describe the often complex shape of these curves with as few parameters as possible. In the case of the statistical distribution models this is done by fitting well known statistical distributions (e.g. a Gaussian, which has three adjustable parameters) to the experimental data. Since there is no a priori assumption about the number and character of binding sites these models easily accommodate non-specific secondary interactions. However, their predictive capacity is limited to interpolations within the range of experimental conditions and they are incompatible with current speciation models.

Another model which does not make *a priori* assumptions about the character and distribution of binding sites is the surface complexation model. However, this concept is only applicable to more or less solid substrates (which may be true for very large organic molecules). Of course, it does not consider explicitly true chemical bonding. Speciation codes do exist which incorporate this type of model. The approach can be further refined adopting concepts of colloid chemistry, namely the formation of micelles, which then can accommodate phenomena of changes in conformation as well.

At present there is no comprehensive model available which includes all the phenomena over the whole range of environmental conditions of interest which have been observed for natural organic matter and which will determine significantly their binding behaviour.

As a result of this review, work has been initiated to incorporate a model for complexation which combines the discrete ligand approach with a modification of the association constants due to electrostatic forces employing surface complexation theory, into PHREEQE. Recommendations have also been made to develop further the random-structure model of fulvic acid.

3.5 FIELD TRACER TESTS

3.5.1 Conservative tracer tests

Three tracer tests have been conducted at re-circulation rates of 2 l/min. An initial test with in-active chloride was compared with a test using ^{131}I and ^{3}H. A sebsequent test was carried out using the Co-EDTA complex. Results show that ^{131}I and Cl move at approximately the same rate but ^{3}H is slightly retarded. Dispersivities were determined using a curve fitting technique based on the 1-D advection dispersion equation with an exponentially decaying source. Logitudinal dispersivities were generally 2 orders of magnitude smaller than the migration distance, at around 4.5×10^{-3} to 1.3×10^{-2} m (Sen and Williams, 1989).

The fastest time to peak breakthrough was 3.25 hours, 3 times faster than that predictedusing the 1-D model. This is due to heterogeneities in the aquifer which necessitates a 2-D model in which the layers are assumed to be 15 cm thick (equivalent to the vertical separation between multi-level sample points).

The information from the hydraulic conductivity profiles has not been used to develop a deterministic model. Rather, the tracer test data has been used to infer hydraulic conductivity profiles of individual layers based on the time to breakthrough. Variations in K needed to create significant flow variations are within an order of magnitude and are probably within the range of accuracy normally achieved for direct hydraulic measurements using falling head tests. The profiles inferred from the tracer test are not well correlated with the directly measured profiles in boreholes slightly off the centre line of the experiment, but are corellated with measurements of hydraulic diffusivity determined in rapid transient response tests using the multi-level samplers fitted with pressure transducers.

The influence of the boreholes, gamma probe access tubes and multi-level samplers has not been assessed using finite element models with a fine grid. It is suspected that little effect would be identified compared with the effects of aquifer heterogeneity.

3.5.2 Tracer tests with anthropogenic organics.

A field test was considered using the natural organic/cobalt complex which had been shown to be mobile but relatively unstable. However, the rate of dissociation of the complex would have required an input of relatively high radio-activity and high concentrations of natural organic material to produce detectable breakthrough.

A field test using Co-EDTA in conjunction with the conservative tracer ^{131}I has been undertaken to assess the effects of anthropogenic organics on radionuclide mobility. EDTA was added to the groundwater at a concentration of approximately 30 mg/l and mixed with a prepared complex of Co-EDTA for 36 hours. Results show that the Co-EDTA complex moves with negligible retardation but also dissociates. Up to 10% of the mobile cobalt is associated with the colloidal phase and further experiments are indicated to assess colloid stability and mobility.

The protocol developed for this test can now be used for other organic or inorganic species.

4. COCO CLUB ACTIVITIES

At the start of the project, BGS was invited to has participate in an interlaboratory comparison excercise to characterise natural organics from a number of sites within the European Community under the auspices of the CEC COCO club. BGS also organised a colloid sampling and characterisation programme at Markham Clinton (Ross et al 1987, Longworth et al 1989) and has participated in a colloid study at Grimsel (Ross and Milodowski,1988). Colloids have been sampled from the Drigg tracer array (Longworth, 1988).

4.1 Characterisation of natural organics

To initiate the interlaboratory comparison excercise a commercially available sodium humate (Aldrich, Na^+) from the Aldrich Chemical Company was obtained, later protonated (Aldrich, H^+) and distributed along with a protonated sample of humic material (Gohy-573,H^+) extracted from groundwater at the Gorleben research site in Germany. In all, characterisation involved the determination of moisture contents, elemental analysis (carbon, hydrogen and nitrogen), metal contents, analysis of functional groups, determination of infra-red absorbtion spectra including Fourier Transform analysis, ultra violet absorbtion E_4/E_6 ratios, and analysis of molecular size distribution by ultrafiltration (Peachey and Williams, 1987, Peachey et al, 1989). Samples of fulvic acid from Drigg, and humic acid from Fanay Augere (France) have also been characterised in similar ways.

4.2 Methods of extracting natural organic material

The means by which organic material is extracted from groundwater is critical if the subsequent characterisation is to be representative. Traditional ways of extracting humic and fulvic acids are aggressive and involve acidification to pH 1 and nuetralisation with alkali. There is evidence to suggest that such techniques irreversibly alter the natural organic material. Several techniques were considered and of these extraction of humic material by sorption onto DEAE-cellulose (di-ethyl-amino-ethyl cellulose) seemed to have minimum impact. The DEAE cellulose is at its optimum efficiency at the natural ionic strength and pH of the water samples.

The resin was added to 25 batches of water, left to stand and then recovered by filtration. The DEAE cellulose was then placed in columns and natural organic material extracted from it by elution with 0.5 M sodium hydroxide. This organic concentrate was then stabilised prior to storage by neutralisation with dilute hydrochloric acid. Attempts were made to obtain humic acid by precipitating from the acidified humic extract (pH1), and standing for several days, but only trace amounts were obtained. The acidified solution was then neutralised and freeze dried. Attempts were made to extract fulvic acid from the freeze dried extract by repeated treatment with methanol followed by rotary evaporation. This technique was promising but insufficient fulvic acid was available for submission as an inter-laboratory standard.

4.3 Sampling and characterisation of colloids

BGS organised the sampling and chracterisation of colloids from the Markham Clinton public water supply borehole in the confined Sherwood Sandstone of North Nottingham which was known to contain elevated concentratioins of uranium. In this study a comparison was made of colloid populations, compositions and size distributions in colloid concentrate samples taken in the field, of a poorly mineralised groundwater taken from fractured Permo-Triassic sandstone. A comparison was made of two methods; pulsed cross flow ultra-filtration and tangential flow ultra-filtration. The actinide isotopic activities associated with the colloids were also determined using isotope dilution/alpha spectrometery. The results from this interlaboratory comparison excercise indicate that (i) no artifacts are produced by either colloid concentration method, although some aggregation of colloidal particles may occur on storage of aqueous colloid concentrates, (ii) good agreement on colloid populations and size distributions is obtained by different laboratories applying scanning electron miscroscopic analysis to cross flow ultrafilters and (iii) no evidencve was found for a significant change in colloid concentration or actinide loading after the groundwater had been stored in platinised aluminium barrels for approximately 6 months.

At the underground laboratory at Grimsel BGS also participated in the study of colloids involving the analyses of membrane and aqueous colloid concentrate samples. This has shown that the concentration of colloidal (≤ 1 μm) particles in Grimsel groundwater is of the order of 10^9 particles/litre, with about 75% of colloids between 3 and 15 nm in diameter. The composition of the colloids is dominated by silicates, with quartz, silica, biotite, muscovite and also Mn/Fe hydroxides, consistent with the mineralogy of the fracture infill material. Evidence of micro-organisms was found in all filter samples, with both bacteria and possibly amoebae and fungi, although it is not clear if these reflect sample contamination or are naturally present in the groundwater.

The colloid population obtained by membrane filtration depends on filter pore size - the numbers of particles recovered increase with decreasing pore size, and sample volume filtered - the population falls with increasing sample volume, reflecting greater aggregation and super-position of particles on the membrane surface, as the number of particles on the filter is increased, particle-size resolution - for the Grimsel samples, a doubling in magnification from 3000 to 6000-fold approximately doubles the numbers of colloids counted; although the resolution is estimated at about 50 nm (0.05 μm) for these magnifications, the increase in population by a factor of four, seen on reducing the filter pore size from 15 to 3 nm, suggests that much smaller particles are being resolved.

Chemical analysis of colloid concentrates prepared from pre-filtered (≤1 μm) groundwater shows very little difference in composition with groundwater, suggesting that experimental artefacts, such as contamination from the filter cartridge or ion exchange with the membrane, are not caused by dia-ultrafiltration. The only element showing a significant increase in concentration in the pre-filtered concentrate is silicon, which would be anticipated from the SEM analyses of colloid filter isolates. In the colloid concentrate prepared from unfiltered groundwater, increases in concentration are seen for Ca, Na, Sr, SO_4, total S, F, Li, B, HCO_3, Cd and Mo, reflecting the significant numbers of larger mafic particulates, such as biotite flakes, which will also be concentrated in this sample.

A colloid sample from Gorleben has also been filtered and colloid populations determined by SEM analysis.

5. REPORTS

Reports

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FINAL REPORT FOR CEC CONTRACT

FI1W/0073/UK

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Natural Analogue Studies of Radionuclide Migration

P J Hooker

Summary

It is now well-recognised that the study of natural geochemical systems can help us to gain a better understanding of the different processes of radionuclide mobilisation, transport and retardation in the far-field, and to provide a means of testing the CHEMVAL thermodynamic database and transport codes which underpin performance assessments of radioactive waste repositories. The British Geological Survey has been carrying out the CEC supported programme of natural analogue studies of radionuclide migration at three sites:

- the sediments of Loch Lomond, Scotland
- the uranium mineralisations at Broubster in Caithness, N Scotland and
- at Needle's Eye near Dalbeattie, SW Scotland.

The three locations are characterised by surficial sedimentary deposits. In various ways these sediments have played host to the transport and retardation of U/Th decay series isotopes, rare earth elements and iodine over time-scales ranging to about 10,000 years ago and over distances up to 100 m or more.

The chief merit of researching natural systems associated with mineralised concentrations of uranium, thorium and rare earth elements is the ease with which concentration gradients accumulated over long periods of time and large distances can be measured. Although the large time-scale and distance aspects make natural analogue studies uniquely valuable over laboratory experiments, there are difficulties and uncertainties in the quantification of initial boundary conditions, in the assumed past groundwater flow rates and in distinguishing the major processes contributing to the elemental distributions.

The different attributes of each site and the approach adopted for defining and measuring the elemental enrichments, migration and retardation are briefly described. The salient features and results are discussed in terms of how the information base has been used in a variety of relevant models.

Introduction

It is important to have a sound understanding of geochemical processes that will affect radionuclide migration over long time-scales and over distances appropriate to the far-field of proposed radioactive waste repositories [1,2,3]. Research into these natural processes is essential for testing the thermodynamic databases and the underlying transport mechanisms used in the computational predictions made for safety assessments [4]. Where mathematical models have been applied in hydrogeology to the prediction of pollution plume characteristics, experience has highlighted the need for proven codes [5]. As well as verifying the inclusion of relevant processes in the mathematical models, it may be necessary to demonstrate and justify the exclusion of processes (e.g. colloidal transport) that may not have significant influences on a particular radionuclide's movement through the far-field. Having reliable background information is crucial, and will be of considerable value in gaining public acceptance of the multi-barrier concept of radioactive waste management. Generic research models that are used to support a risk assessment for a future repository site can be tested against data and measurements belonging to natural geochemical systems. By studying such systems, we improve our understanding of likely radionuclide behaviour in the far-field and build a firmer foundation on which to increase our confidence in predictive modelling.

Objectives

The objectives are as follows, with the first one relying heavily on the other two:
1. To test the applicabilities of thermodynamic databases in particular CHEMVAL [6], and research codes such as PHREEQE [7], CHEMTARD [8], METIS [9] and STELE [10], for modelling natural systems and the far-fields of radioactive waste repositories.
2. To understand the geochemistry and migration rates of uranium and thorium decay series elements, rare earth elements (REE) and iodine in different sediment environments.
3. To study the processes involved in their geochemical and hydrochemical distributions and to devise ways of measuring their speciation in the solid and water phases.

The studies aimed at these goals have been biased towards attempts to understand in particular the transport behaviour of natural decay series radioisotopes in a variety of low temperature sedimentary environments established since the last ice age. The chief merit of researching natural surface systems associated with mineralised concentrations of uranium, thorium and rare earth elements is the ease with which concentration gradients accumulated over long periods of time and large distances can be measured. Being able to distinguish the process(es) that had governed the distribution of e.g. uranium or iodine, in a sediment over a given period of time was a major reason for investigating the natural analogue sites described below. Following a preliminary reconnaissance [11], the CEC have supported investigations at three sites (Fig. 1):

- Sediments in **Loch Lomond**, Scotland;
- Uranium mineralisation at **Needle's Eye**, Southwick, on the Solway Firth coast near Dalbeattie, southwest Scotland;
- A uranium anomaly at **Broubster**, Caithness, northern Scotland.

To assist in its research programme over the past three years, the British Geological Survey has collaborated with the Scottish Universities Research and Reactor Centre (SURRC) at East Kilbride, with the UKAEA Harwell Laboratory, with WS Atkins Engineering Sciences and, under the auspices of the CEC (cost-shared) Mirage II Project, with the Centre d'Informatique Géologique of the Ecole des Mines de Paris (EMP), Fontainebleau. The studies at SURRC involve (a) an investigation of the speciation of the halogen elements in Loch Lomond sediments and (b) the application of natural decay series analyses to the rocks and waters of the Needle's Eye site. The research carried out at Harwell focuses on a U/Th decay series description of the waters and sediments from Broubster. WS Atkins have carried out modelling studies for the Broubster site, while EMP have modelled the Needle's Eye site.

Approach

In the studies at Broubster and Needle's Eye, a general approach has been followed which consists of trying to define the groundwater flow regime affecting the rock volume hosting the mineralisation or concentration band acting as a source-term and the sink area; this has been achieved by consideration of the regional hydrogeology and by surveying in simple piezometer tubes into the soft sediments at discrete points on a grid system to measure hydraulic conductivities and gradients. These same piezometers served to sample the groundwaters carrying the transported species. In situ pH and Eh were measured via a flow-through cell; in some cases it was necessary to exclude air with nitrogen to prevent oxidation of Fe(II) and sulphide ions. Standard methods e.g. titration, inductively coupled plasma optical emission spectrometry (ICPOES), inductively coupled plasma mass spectrometry (ICPMS) and ion chromatography (IC), were employed for the chemical analysis of the waters. Extensive suites of samples were collected for mineralogical investigation by means of X-ray fluorescence spectrometry (XRF), autoradiography, fission track registration and subsequent scanning electron microscope (SEM) analysis. Gamma and alpha spectrometry methods were applied for U/Th decay series analysis of selected samples. The delayed neutron activation method was also applied for U. The solid phase concentration gradients were defined in 3-D as far as possible and matched with flows and groundwater abundances.

The approach has involved:

(i) the development and application of field and laboratory techniques for determining the chemical concentrations and forms of U, Th and I and Br in groundwaters; this has included the examination of colloids;

(ii) an attempt to discover the role of natural organic compounds in the geochemical distributions of the uranium and thorium decay series radioisotopes;

(iii) the interpretation of hydrogeochemistry data with equilibrium speciation codes such as PHREEQE and CHIMERE [10], and to test the thermodynamic databases, especially CHEMVAL, employed in the codes;

(iv) the determination of groundwater flows and U/Th distributions to make comparisons with the outcomes of modelling with coupled chemical transport codes such as CHEMTARD and STELE.

Site attributes

It is not the intention to give detailed descriptions of each site investigated as these are given in the technical reports quoted; instead the salient characteristics and findings will be presented.

(1) The Loch Lomond sediments

These consist of a marine horizon, approximately one metre thick, underlain and covered by freshwater deposits that have accumulated a thickness of about 3m since the retreat of the sea around 5400 years ago [12]. Previous studies of the sediments from the point of view of diffusion of Br, I, U and Ra from the organically associated enrichments in the marine layer into the overlying deposits [13,14,15] have led to further sampling and the development by SURRC of a new sensitive low-blank technique for analysing halogens in 0.1 ml of pore water [16]. The consequent concentration-depth profiles for chloride, bromide and iodide [17] are being interpreted and modelled in terms of diffusion and release rates from the marine horizon enrichments [18]. Most of the halogen profiles can be explained in terms of diffusion without retardation.

(2) The Needle's Eye site

The ancient cliff at Needle's Eye on the Solway Firth coast contains leachable pitchblende within polymetallic-carbonate breccia veins which have acted as a source of soluble uranium found dispersed in the post-glacial mudflats stretching below the cliff to a tidal creek [19,20]. These estuarine and intertidal silt deposits known as the Merse were laid down in the past 10,000 years or so upon bedrock composed of Silurian hornfels siltstone faulted against Carboniferous limestone (Fig. 2). A wide range of soil, sediment and rock samples were collected for chemical and radiometric analysis. Groundwaters collected from a grid system of piezometers established within the sedimentary cover, from the streams and from the cliff, were analysed for their chemical compositions. At the same time a series of simple hydrogeological tests were conducted to ascertain the flow characteristics through the deposits [21]. A pit section through these sediments has provided detailed samples for comprehensive analysis by XRF and neutron activation techniques; high resolution gamma spectrometry has contributed information on the distributions and disequilibria between uranium and its daughter radioisotopes [22]. Alpha spectrometry results have been obtained on a wide range of samples [23]. The U/Th series data have been interpreted to derive loss rate estimates of 5 x 10^{-5} to 10^{-6} per year for U from the cliff source area to the Merse sediments, depending on the time-scale assumed, and a halving distance of 10 m for surface flow of U towards the tidal creek [24].

Integration of autoradiographic and fission track techniques has been used to locate and quantify U in samples of soils and silts proving that U is significantly retarded by organic matter [25]. Accumulation occurs at two distinct levels in the sediment profile, at around 50 cm depth and between 100-150 cm. Fission track registration indicates that at the 50 cm level uranium is located within the fine organic matter of the peaty matrix and is strongly enhanced around open root channels where concentrations reach several hundred ppm. This suggests fixation due to reduction. In the deeper accumulation, U is located almost exclusively within plant roots, and is particularly concentrated (often >1000 ppm) within specific cellular structures related to water transport. In addition, U-As-Cu and Bi bearing minerals found on root surfaces indicate that metal-fixing fungi may be important in arresting mobile uranium.

These data have been combined with analyses from groundwaters sampling various levels of the deposits to produce a geochemical model of the U distributions and transport mechanisms. There are thought to be two main inputs of dissolved uranium(VI) into this sediment system, surface flow of groundwater from the exposures of the mineralisation in the cliff, and upward flow from the bedrock below. The fixation of uranium in the sediments is controlled by organic matter in the upper humic layers of the section, and by organic matter (roots) and associated iron oxyhydroxides in the deeper silts. This concentration of U in the silt is divorced from the sub ^{234}U daughters. In contrast, thorium is coherently associated with its daughters within detrital resistate minerals.

Based on numerical simulations with the METIS flow code, a hydrogeological framework was established for the region and for the restricted area of study at the base of the cliff [26]. Preliminary hydrogeochemical modelling was carried out with the groundwater composition data using the CHIMERE program (which is similar to the PHREEQE equilibrium code) in conjunction with the second version of the CHEMVAL thermodynamic database [27]. It was possible to classify the groundwaters into three distinct groups on the basis of their ionic strengths. Weakly mineralised water indicated an origin from the hornfelsed siltstone of the cliff, mainly by surface run-off. Strongly mineralised groundwaters reflected (a) the influence of components dissolved from the limestone bedrock south of the E-W fault, and (b) the influence of tidal contributions; such waters occurred mainly in the Merse sediments south of the fault. Moderately mineralised waters indicated a siltstone origin with some marine influence.

The calculation of saturation indices of selected minerals in contact with the groundwaters along a N-S transect and at varying depths allowed the deduction of flow paths and an indication of the various rock types through which the water had travelled and interacted. Piezometer waters from the area south of the fault showed oversaturation or equilibrium with calcite demonstrating that there must be a vertically ascending component of flow from the limestone substratum. Field measurements had independently confirmed this deduction. In the sediment area north of the fault where the groundwaters are undersaturated with respect to calcite, the calculations were compatible with the groundwaters being in equilibrium with nahcolite [$NaHCO_3$], an improbable phase in such a geological environment.

The controls governing uranium solubility were modelled on the basis of UO_2 and liebigite [$Ca_2(UO_2)(CO_3)_3.10H_2O$] saturation indices. It is possible to reproduce the uranium concentrations in the groundwaters using the solubility of liebigite, but this U(VI) mineral has not been found in the sediments. A better approach involves the idea of reduction-precipitation of U(VI) to U(IV) as uraninite Uranium concentrations in the groundwaters and sediments contained in the section nearest the cliff were simulated using the coupled code STELE [28]. The mobile species are U(V1)-carbonate complexes which are stable at +400 mV, pH 7.8 and CO_2 pressure of $10^{-3.2}$ at. On contact with the organic-rich sediments with a lower Eh of +250 mV, U(V1) is reduced and uraninite can precipitate with a rate constant of 1 d^{-1}; this produces a steady state after a month. The calculated aqueous U concentrations agree well with the measured values. Also the computed accumulation of U in the sediments over 5000 years is 500 ppm, not much different from observed concentrations. The model has predicted the overall effects observed by measurement. Whether this is a satisfactory outcome when the fission track registration evidence points to organic matter being directly responsible for the fixation of uranium is open to debate.

It was not possible to reproduce the observed 2-D distributions of U with METIS and the Kd concept using laboratory derived K_d values [29]; in this model the groundwater U concentration declines too slowly from the input concentration of $4x10^{-7}$ M, and gives a pessimistic prediction for U transport [28].

Despite the limited hydrochemical data, the uncertainties in the redox potentials and the limited understanding of the role of organic matter in fixing U, much has been revealed by this thermodynamic equilibrium approach to modelling the groundwaters. It has indicated potential rock sources for the dissolved components, confirmed likely flow paths and provided ideas for coupled transport modelling. On the negative side, this approach cannot deal with disequilibrium and is inadequately prepared for describing the role of organics towards uranium movement in the sediments.

(3) The Broubster site

The site is divided into two areas by a single-track road running north-south. To the west is the source area for uranium, a laminated limestone of the Caithness Flagstone Group belonging to the Orcadian lake sediments of the Middle Old Red Sandstone. Here, the area has been quarried on a small scale in the last century, and backfilled with quarry spoil. Approximately 100 m away downslope east of the road is a peat bog (<10,000 years old), underlain by boulder clay and the siltstone bedrock. This is the sink for uranium transported by groundwater flows along divergent paths from the source area (Fig. 3). The occurrence of anomalously high uranium (0.1 wt%) in the peat soils of Broubster was described in 1972 [30]. Recent field work [31] at the site had the objectives of:

1. Assessing the hydrogeology
2. Collecting and describing samples of the 'source-term' uraniferous mineralisation
3. Collecting groundwaters for chemical analysis, for U/Th speciation measurements with a portable chromatographic separation kit [32], and for U/Th series determinations at UKAEA
4. Collecting samples of the peat (sink-term) for metal analysis, C-14 dating and organic characterisation work [33].

A major source-term has been identified as a thinly laminated limestone approximately 40 cm thick at a depth of 90-130 cm. The lamellae consist of repeated units often less than 1mm thick, built up of a micrite (or algal carbonate mud) band and an organic silt band. The siltstone lamellae consist essentially of detrital quartz plus minor K-feldspar, plagioclase, muscovite, chlorite, biotite and mudstone pellets. The quartz and feldspars have developed euhedral overgrowths which cement the laminae. Authigenic pyrite is common as very fine octahedra or framboidal aggregates. Organic material is intermixed with this detrital component. Concentrated by biological and mineralogical processes the limestone is hence considerably enriched in uranium with up to 30 ppm U compared with the regional siltstone lithologies of 2-7 ppm U. The principal hosts for uranium in the limestone horizon are the carbonate bands, which comprise 80% of the whole rock with an evenly dispersed U concentration of 25 ppm, and the minor organic-silt layers with up to 60 ppm U. Hydrocarbon contained within the silt layers enclose crystalline phosphate/titanate minerals which have retained U. It is important to note too that a carbonaceous fossil fish fragment found within an organic silt layer had much U associated with it and had remained in place unaffected for 400 Ma [34].

Recent to present day weathering processes are effective in mobilising uranium. Oxidation of pyrite is of significance, causing acid leaching. Dissolution of calcite by percolating groundwaters also provide direct release of uranium. Little evidence exists both from mineralogical and U/Th series work for Th mobilisation. However, rare earth element mobilisation is significant as evidenced by the REE patterns [31]. Once in solution, complexed with carbonate, uranium passes out of the quarried area via divergent flow paths. Uranium is removed from solution by complexing principally with organic matter within the waterlogged peat deposits. Iron sesquioxides are also important, acting as scavengers of uranium.

Experimental measurements of groundwaters for U and Th using the portable chromatographic separation kit and ICPMS analysis indicate a soluble humate complex of U(VI) that is sorbed onto the peat. Modelling studies with PHREEQE and CHEMTARD [35,36], attempted to predict the speciation, retention and transport mechanisms for the actinide elements. Used in conjunction with the REE distribution data and complementary experimental measurements the work has led to a coherent interpretation of U, Th and REE behaviour at the site. The applicability of equilibrium codes and thermodynamic data in the field situation at Broubster has been demonstrated, but the inadequacies in modelling organic-metal interactions have again been highlighted. The preliminary 1-D transport modelling has also served to highlight further geochemical and hydrologic data required to characterise the system more thoroughly. Although the computations are currently semi-quantitative because of the absence of this information, a number of conclusions can be drawn. Thus, in the case of uranium it is apparent that:

- oxidation-reduction reactions do not necessarily control U fixation
- U levels in soils are not solubility constrained
- carbonates (and possibly phosphates) dominate the inorganic speciation of U
- organic complexation becomes dominant below pH 6
- U is present in 'true' solution and thus amenable to simulation by aqueous speciation models.

The above results compare well with experiment and concur with the known distribution of uranium at the site. For thorium, the situation is less clear owing to probable co-precipitation of Th and Fe hydrous oxides and the likely presence of Th in colloidal form. Further U/Th series experimental work [37], including ultra-filtration, has confirmed that colloids are moving Th through the system; also, whereas the bulk of the ^{238}U and ^{234}U activities in the peat are associated with the organic matter component, there is evidence for a repartition of the daughter ^{230}Th away from the organic phase into amorphous iron oxyhydroxides. The REE information is awaiting detailed modelling treatment.

Surface diffusion in clay

In addition to these site investigations, a desk study of surface diffusion and mass transport of cations in clay was carried out [38]. The central finding was that a better definition of 'effective' porosity would greatly improve our understanding of solute transport mechanisms.

Major findings and conclusions from studying the UK natural analogue systems

The studies have led to the development of some improved chromatographic separation techniques for measuring the speciation of the halogens and uranium in groundwaters.

The combined use of autoradiography, fission-track registration and SEM techniques has proved a powerful way of examining the geochemical forms of U and Th in the solid phase. Without knowledge of speciation in both phases, it is difficult to determine the active mechanisms affecting distributions, and modelling becomes merely a parameter fitting exercise.

U/Th decay series studies of natural geochemical systems have been essential in tracing colloids, defining degrees of disequilibrium and chemical fractionation, describing the long-term partitions of the radioisotopes between various phases, and in deriving migration rates for particular radioisotopes. The migration rates of Th isotopes from mineralised concentrations are orders of magnitude smaller than for U.

The emphasis in testing the CHEMVAL database and in modelling has been focused towards the actinide elements. There is increasing evidence that colloidal migration of U is far less important than transport in true solution. In the case of Th, however, colloids (both inorganic and organic) may constitute a major proportion of the total speciation.

The K_d concept is inadequate to describe the distribution of uranium in the geosphere. This is probably true of other redox sensitive elements. Laboratory batch sorption measurements are not relevant to long-term migration for uranium in the far-field.

The successful application of surface complexation theory [39] in coupled chemical transport codes like CHEMTARD should be reinforced by more laboratory measurements of the appropriate parameters.

Organic matter, both dead and living, has been observed to play a pivotal role both directly and indirectly in the retardation of uranium. Peat and organic matter in silts are very efficient at retarding and fixing dissolved U. The capability of the relevant codes to deal with metal-organic matter interactions is poor [40].

Nevertheless, the relevant codes (PHREEQE, CHIMERE, METIS, CHEMTARD and STELE) have been used to simulate the overall effects of U migration studied at the sites, and their applicability to the natural analogue sites has been shown to be appropriate within limitations.

General points

Collecting and modelling data in isolation of the nature of the problems and without regard to the context has been avoided, but the interactive two-way exchanges of information between mathematical modellers and experimenters has only been possible since the establishment of the conceptual models. These took longer to establish than had been expected at the start.

More thorough preliminary surveys of the Needle's Eye and Broubster sites at the beginning by means of geophysical and hydrogeological techniques would have speeded the iterative process now in action between data collection and modelling.

The studies have highlighted the uncertainties inherent in natural geochemical systems [41, 42].The uncertainties revealed in these natural analogue studies are no different from those to be encountered in a site investigation for a repository. The confidence value and success of applying tested codes for calculating radionuclide transit times in performance assessments may depend on comparisons with the geo-releases of natural radioactivity weathered from the host rocks of the repository into the accessible environment [43].

Future areas of work

It has been demonstrated that the findings of natural system studies give a crucial insight into future model development and performance assessments of proposed radioactive waste disposal sites. For this reason further collaborative analogue studies are continuing.

The emphasis on relating experimental observations with speciation and transport modelling will be developed further. Rock, sediment and water samples from the localities at Needle's Eye and especially Broubster (being close to the proposed site at Dounreay for an investigation into its suitability for a deep repository), will continue to be analysed in order to describe the geochemical distributions of U/Th decay series elements in the sources and sinks, and to model the governing mechanisms. The application and development of techniques to ascertain aqueous speciation both inorganic and organic will be pursued. Relating the results to the in situ redox conditions will be of particular interest. The subsequent fate of a metal retarded in an organic matter complex or as a surface complex on amorphous iron oxide, and the kinetics of the transformations into more permanent crystalline lattice structures are poorly known; these areas warrant further study.

Focusing on obtaining migration rates of different U/Th species under similar environmental conditions of erosion and weathering will place important constraints on natural releases into the biological system and hence the food chain. More attention will be paid to ^{226}Ra which always appears in lists of radionuclides that contribute high dose risks long after closure of the repository. The sites at Needle's Eye and Broubster are ideal for directing attention at the different weathering rates of the U/Th decay series elements.

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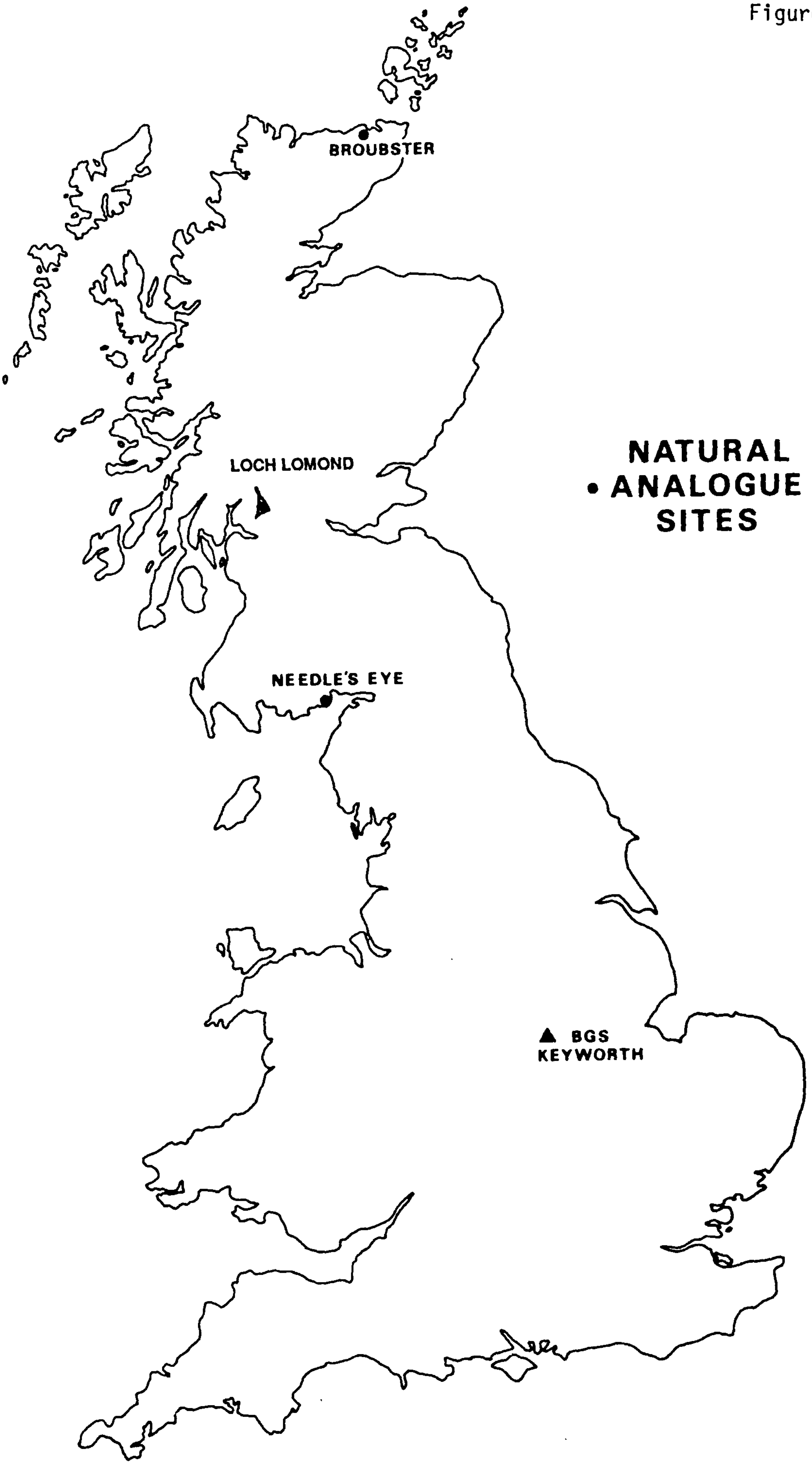
Figure captions

Fig. 1. Location map for the natural analogue sites in Britain.

Fig. 2. A schematic N-S section of the Needle's Eye site illustrating the main flow paths; the distance from the creek to the cliff is about 80 m; the cliff is about 35 m high.

Fig. 3. A schematic section (100 m long) of the Broubster site, showing the main flow paths.

Figure 1


NATURAL
• ANALOGUE
SITES
BROUBSTER
LOCH LOMOND
NEEDLE'S EYE
▲ BGS
KEYWORTH

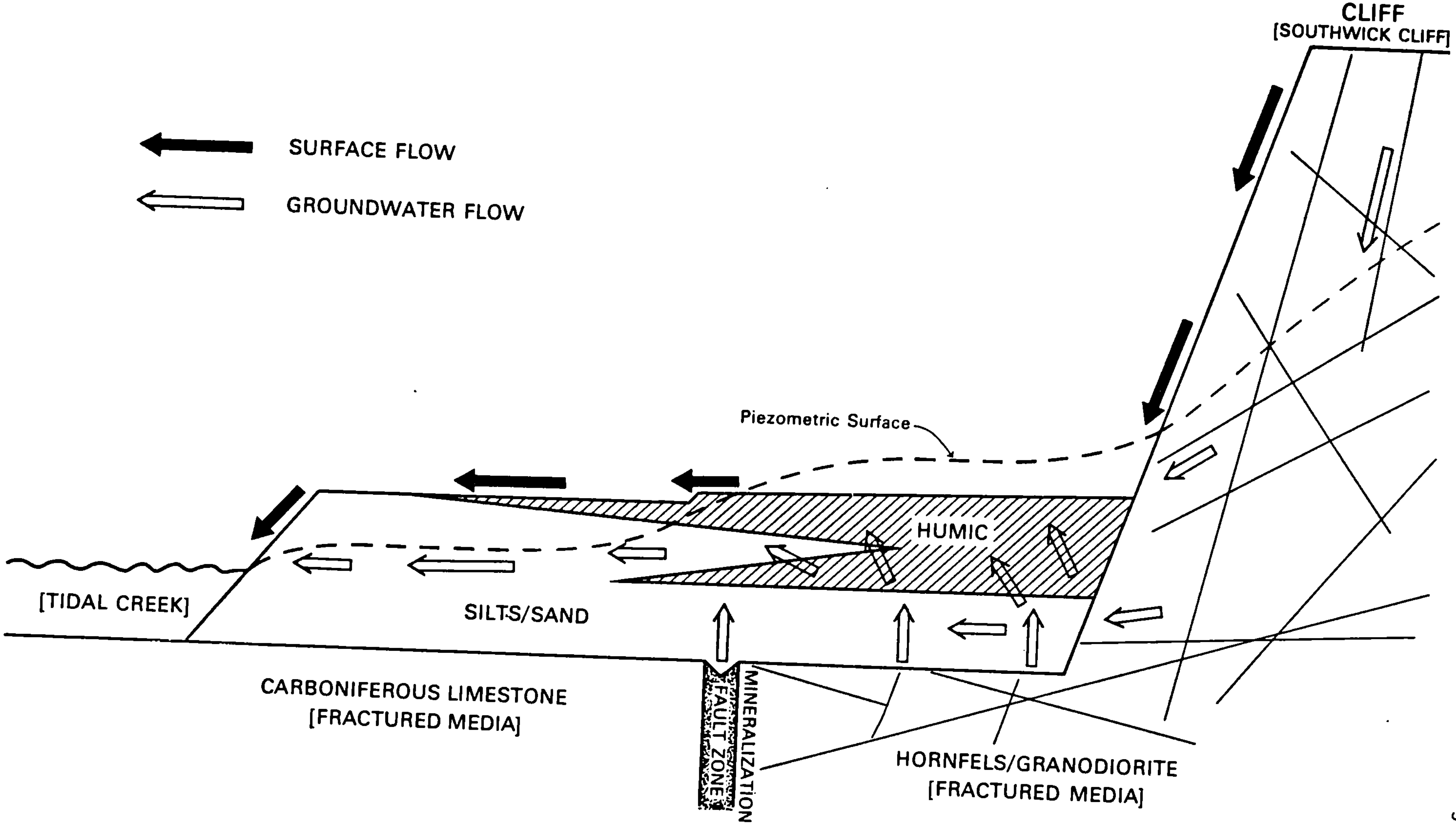

CLIFF
[SOUTHWICK CLIFF]
SURFACE FLOW
GROUNDWATER FLOW
Piezometric Surface
HUMIC
[TIDAL CREEK]
SILTS/SAND
CARBONIFEROUS LIMESTONE
[FRACTURED MEDIA]
MINERALIZATION
FAULT ZONE
HORNFELS/GRANODIORITE
[FRACTURED MEDIA]

Figure 2

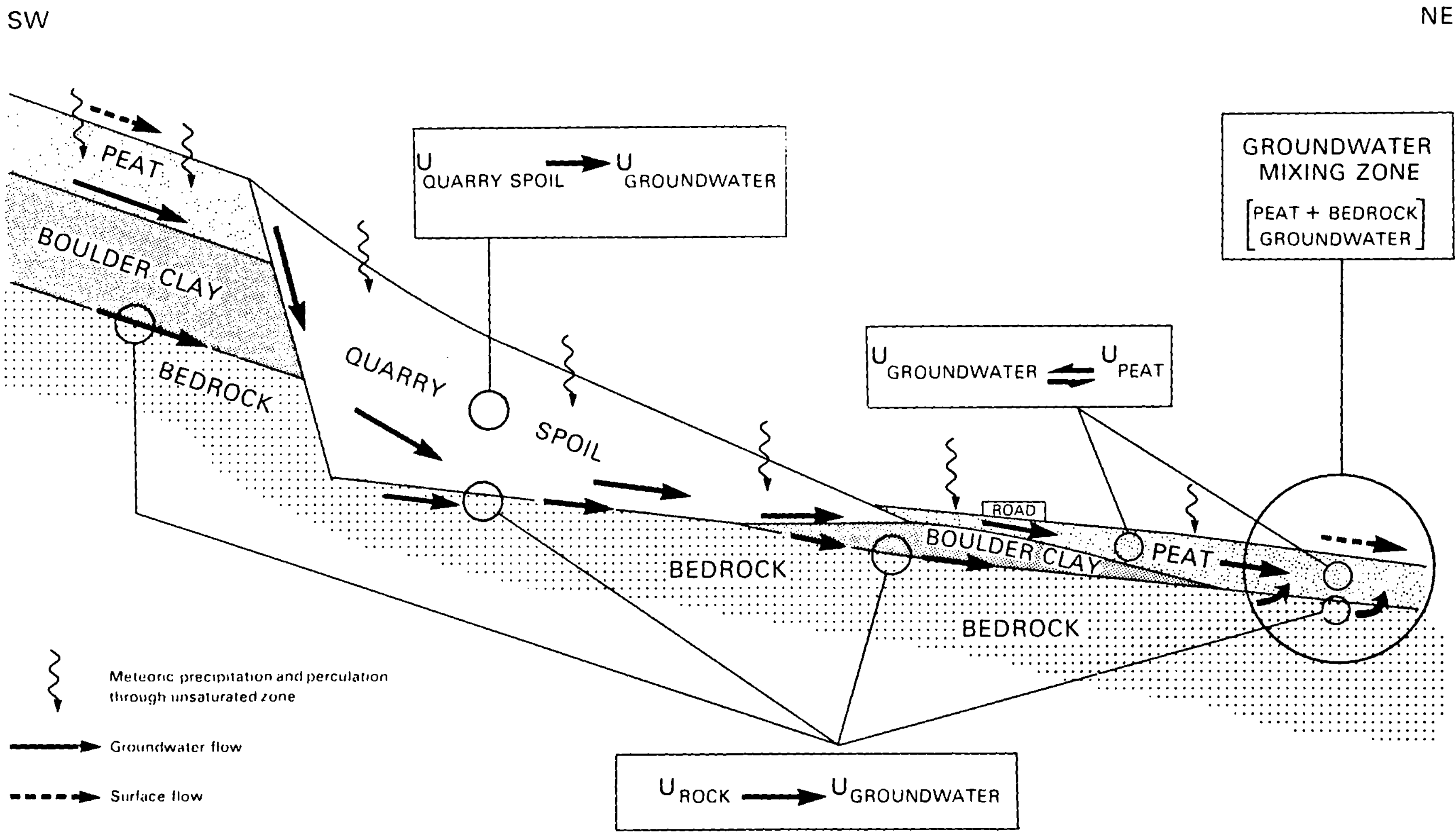

SW
NE
PEAT
BOULDER CLAY
BEDROCK
QUARRY
SPOIL
BEDROCK
ROAD
BOULDER CLAY
PEAT
BEDROCK
U QUARRY SPOIL → U GROUNDWATER
GROUNDWATER MIXING ZONE
[PEAT + BEDROCK GROUNDWATER]
U GROUNDWATER ⇌ U PEAT
U ROCK → U GROUNDWATER
Meteoric precipitation and perculation through unsaturated zone
Groundwater flow
Surface flow

Figure 3

FINAL REPORT FOR CEC CONTRACT

FI1W/0085/UK

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Faults in clays: their detection and characterisation

M. A. Brightman

1 Introduction

The British Geological Survey, in cooperation with ISMES of Italy, has carried out a research programme into the properties of faults cutting clay formations. Clays are being considered as potential hosts for radioactive waste repositories in a number of countries. Two physical properties of clays have stimulated this interest. Firstly clays generally have very low hydraulic conductivities and consequently small advective groundwater flows; secondly clay minerals are considered to sorb radionuclides strongly. Faults are known to occur in most sedimentary environments, but the extent to which such faults change the physical properties of the sediments, and the clays in particular, is poorly understood. Potentially, faults could act as near vertical conduits for direct groundwater flow from the repository zone to the surface, effectively forming a short circuit in a larger, less active, regional groundwater flow system. The role of clays as barriers to fluid migration also has important implications for hydrocarbon exploration because the sealing properties of clay cap rocks may be reduced or enhanced due to faulting.

In the UK clay formations generally form low lying vales with very little topographic variation, the clay often being covered with a thin layer of post-glacial sediment. Where the clay is exposed deep weathering tends to produce a lithology of uniform appearance regardless of original subtle lithological variations. The combination of these factors means faults in clay terrains are very difficult to locate by conventional geological mapping. This research project was therefore initiated with two major aims. Firstly to develop geophysical techniques to detect faults in clay lithologies and to measure their geophysical properties. Secondly to measure the hydrogeological properties of faults in clays and to determine their hydrogeological effect on the groundwater flow regime of a sequence of clays and aquifers.

2 Selection of potential research sites

The first stage was to carry out a desk study to identify potential research sites (Alexander, 1989) and then to select two sites for reconnaissance geophysical surveys. The geophysical and hydrogeological characteristion of a fault would be optimised if the site was relatively simple and met a number of geophysical and hydrogeological pre-conditions. A conceptual description of the 'ideal' fault research site was therefore drawn up which could be compared to the features of sites identified by the desk study. This 'ideal' site should have mudrocks at surface with no superficial deposits. These mudrocks should have a minimum thickness of 30 metres on the upthrow side, and a maximum of 80 metres on the downthrow side. The mudrocks should be underlain by an aquifer 10-20m thick, which is in turn underlain by more mudrocks. The minimum displacement of the fault should be 10 metres. The fault should have a well defined fault plane or zone. The underlying aquifer should be artesian giving rise to upward groundwater flow through the fault. There should be good geological, geophysical and hydrogeological data available in the area for background control purposes. The site should be remote from built up areas but still be easily accessible.

A list of ten potential sites was drawn up and all the readily available data collected and compared to criteria derived from the 'ideal' site. None of the sites matched the ideal site exactly but two sites were clearly more suitable than the others. These were two disused airfields from the second world war, at Wombleton in the Vale of Pickering, North Yorkshire and at Down Ampney in the Thames Valley of Gloucestershire, and these sites were selected for reconnaissance geophysical surveys. At Wombleton Kimmeridge Clay overlies a 20m thick Corallian sequence of limestones and sandstones. At Down Ampney Oxford Clay overlies the Kellaways Beds, a series of silty and clayey sands and clays, which in turn overlie the target aquifer, the Cornbrash limestone.

3 Reconnaissance geophysical surveys

One important objective of the project was to establish an efficient geophysical methodology for the detection and delineation of faults in clays and the measurement of their displacements, to an accuracy adequate for their detailed investigation by borehole drilling. The geophysical reconnaisance performed at Down Ampney and Wombleton (Jackson et al, 1989a) fell into two parts:-

1. The appraisal of the sites to allow final site selection, together with an initial evaluation of the proposed geophysical techniques.
2. An extensive survey of the Down Ampney site to select the general drilling location, with a refinement of the geophysical methodology.

Faulting which causes the displacement of plane interfaces between two regions of differing physical properties may be detected by geophysical techniques. The techniques should be sensitive to the property changes as part of a three-dimentional geophysical target. For example a very small relative displacement of an interface may not be detectable even though there is an order of magnitude change in properties. The geological setting at both Down

Ampney and Wombleton indicated a relatively large change in clay thickness, with the expected throw of the fault equal to or greater than the thickness of clay on the upthrown side. These relatively large throws could be detected by both seismic and electrical resistivity methods because the contrast in properties is very large in both cases.

The primary geophysical technique, Schlumberger resistivity traversing, modified as necessary, was selected on the grounds of high lateral resolution, logistical efficiency and its capability of almost immediate field interpretation and appreciation as the survey progressed. Schlumberger resistivity sounding, shallow seismic refraction and electromagnetic profiling, were available as secondary techniques.

For each site a profile of anticipated geological formations, thicknesses and resistivities was compiled. From these, computer models were generated to estimate the composite field resistivity response ('apparent resistivity') for the minimum and maximum clay thicknesses anticipated at each site. This confirmed that at both sites a resistivity spread length ('AB') of approximately 300m should maximize the resistivity contrast from a probable fault and be insensitive to variations in any superficial formations. Graphs were also prepared for the instant conversion of field readings to approximate clay thicknesses.

For the appraisal survey at Down Ampney, the main traverse line of c.1500 metres was orientated across the anticipated direction of faulting, at each end terminating near an existing borehole. The resistivity traverse revealed a very marked anomaly, coincident with an abrupt change in clay thickness from about 30 to 50 metres, which was confirmed by seismic refraction. A less marked anomaly was detected by electromagnetic profiling at the same location. A second parallel line, alongside the perimeter road of the disused airfield was degraded by erratic readings which are consistent with buried objects near the electrodes. It was concluded that the buried remains of the airfield infrastructure caused the erratic behaviour making this part of the airfield unsuitable for further surveys.

Similar problems were encountered during the appraisal survey at Wombleton, but in this case their effect was much more serious as they affected all the traverse lines attempted in the western half of the site. It is probable that a major fault crosses this site, although its precise location could only be confirmed with certainty along one traverse. The resistivity sounding data indicated that the thickness of clay on the down throw side could be well in excess of 100m. It was concluded that geophysical surveying was likely to be far more problematic at Wombleton than Down Ampney and Down Ampney was selected as the main research site. Other factors which contributed to the abandonment of Wombleton in favour of Down

Ampney included the much greater tectonic complexity of the area, the greater variability of the Kimmeridge Clay and the relatively sparse availability of geological data.

During the extensive main survey of the Down Ampney site over 7km of resistivity traverse were surveyed, covering an area of about 1.5km^2. The fault detected in the earlier survey was intersected by six traverses and found to have a slightly sinuous E-W trace (Figure 1). The strata to each side of the fault appear to have only gentle flexures and dips and to be devoid of further faulting, with one possible exception at the northern extremity of the site. On both the southern (upthrow) and northern (downthrow) sides of the fault the strata appear to dip to the east, more steeply in the latter case. As a result the throw of the fault appears to increase eastwards.

Figure 2 shows the shape of the resistivity traverse plots which are particularly striking as they cross the fault. As the fault is approached from the upthrow side, the resistivity rises quite steeply to a peak, the magnitude of which decreases from west to east, and then plummets extremely steeply to a shallow trough, from which it rises rather irregularly to the down throw plateau. The fault has been nominally taken at the mid point of the steep fall, which can be located on the ground consistently to a precision of just a few metres. Numerical modelling suggests that this signature may be due to a combination of the fault displacement and a narrow steeply dipping zone of markedly different resistivity in the clay.

Whilst the plan location of the fault appears to be of unexpectedly high precision, the interpretation of the resistivity data in terms of thicknesses and depths seems distinctly more uncertain. Several of the traverse lines were surveyed, in whole or in part, with a reduced spread length of 200 metres. After applying the appropriate conversion factors, the resultant resistivities differ markedly, but very consistently, from the data obtained with the 300 metres spread length. This strongly points to the original model profile of thicknesses and resistivities being in error. Conventional interpretation of the three resistivity soundings can be achieved only by major alterations to the layer thicknesses. As the thicknesses are very well controlled by boreholes in the immediate vicinity, it would appear that the conventional assumption of isotropic resistivities may be erroneous. This hypothosis was investigated at a later stage of the project, and the resistivity anisotropy of the clays at Down Ampney was demonstrated to be between 2 and 3, see section 6 below. The depths deduced from the resistivity surveys could only be regarded as approximate and required confirmation by drilling.

4 Borehole drilling at Down Ampney

The borehole drilling at Down Ampney fell into two stages, firstly, a series of exploratory boreholes and secondly the drilling of a measurement array (Brightman et al, 1989a). This was carried out at the eastern side of the airfield where the resistivity surveys indicated the clay thicknesses to be greatest.

The aims of the exploratory phase of drilling were twofold. Firstly, by drilling a borehole on each side of the fault, to establish lithostratigraphic and geophysical reference sections through the clay strata down to the top of the Cornbrash, and thence deduce the throw of the fault. Secondly, to locate the fault as accurately as possible from a tranverse line of shallow boreholes.

The nine exploratory boreholes (DA1-DA9) were fully cored to provide the reference sections and core samples for laboratory testing, and their locations are shown in Figure 3. The two deep reference boreholes (DA1 and DA2) were sited outside any conceivable fault influence in order to determine the fault throw. The initial intention of the shallow exploratory programme was to drill a traverse of boreholes at spacings of about 10m. In each borehole about 10m of unweathered Oxford Clay would be cored and a natural gamma log run. The lithostratigraphy of these cores and the gamma logs were to be compared to the reference boreholes. From the comparisons the approximate location of the fault and the extent of any disturbed zone would be deduced. If the fault was discrete and near vertical, the probability of intersecting it with shallow boreholes would be small. The four boreholes DA3 to DA6 were each drilled to about 20m, through sequences essentially similar to DA1, with no obvious evidence of any fault associated features. However in DA7 progressively more steeply inclined strata were encountered until the main fault zone itself was reached. Beneath this the dips rapidly decreased and the borehole was terminated in undisturbed Cornbrash. DA8 was drilled at a nominal distance of 2m from DA7 in order to obtain a preliminary estimate of the dip of the fault.

The purpose of the exploratory boreholes was to prove the existence of the fault and determine its location; consequently very little effort was put into completing the boreholes for future measurements and they were completed by simply inserting plastic casing and screen without any grout.

The aim of the measurement array phase of drilling was to produce an array of boreholes straddling the fault in which the geophysical and hydrogeological evaluation of the fault would be carried out. This line of thirteen boreholes (DA10-DA22) was sited 30m away from the exploratory boreholes (Figure 3) to minimise the effects of hydraulic connections via the annuli of the poorly completed exploratory boreholes. The boreholes were partially cored for the dual purposes of providing core samples for laboratory tests and to allow the boreholes to be located accurately with respect to the fault zone. The boreholes fell into two categories; firstly narrow diameter piezometers and secondly cased and screened boreholes of 100mm nominal diameter (Figure 4). The piezometers form a series of hydraulic observation points in the clays and are grouped into three nests, one each to either side of the fault and the third in the fault zone itself. The four larger diameter boreholes were for cross-hole seismic tomography and hydraulic pumping tests. Two are positioned remote from the fault, to either side, with screened sections in the Cornbrash. The other two penetrate the fault zone with screened sections in the Cornbrash and the Great Oolite.

The measurement boreholes were a specifically designed array of holes carefully located for geophysical and hydrogeological measurements. The successful completion of the boreholes was a crucial component of the project. The piezometers into the low hydraulic conductivity Oxford Clay needed high quality bentonite and grout seals to allow accurate measurements of hydraulic conductivity and groundwater head. The grout needed to be of sufficient rigidity to allow the transmission of seismic s-waves and be free of voids in the anulus.

5 Geological structure of the Down Ampney Fault

This section briefly describes the various lithological units at Down Ampney and the structure and geometry of the fault. The core samples recovered from the exploratory boreholes were logged in detail and the geological sequence proved by the boreholes was described by Horton et al (1989) and the findings of the second phase of boreholes are summarised by Brightman et al (1989a).

The geological sequence penetrated at the Down Ampney site extends from the Middle Oxford Clay at surface down to the Great Oolite Group, and a simplified geological section is shown in Figure 5.

The Middle Oxford Clay Member comprises pale grey, slightly calcareous mudstone with subordinate silty mudstones and rare weakly calcareous siltstones. The member is generally poorly fossiliferous with a macrofauna dominated by small (mainly pyritised) ammonites, scattered bivalves and small gastropods. However, it is also characterised by slightly greenish grey, richly fossiliferous mudstones packed with the small, thin shelled bivalve Bositra buchii (Roemer). The Lower Oxford Clay Member consists largely of mudstone with subordinate thin silts, siltstones and fine sandstones in the lower part. A thin argillaceous limestone, the Acutistriatum Band, occurs in the upper part, and at least two septarian nodule horizons in the lower part.

The Kellaways Formation can be divided into an upper Kellaways Sand Member and a lower Kellaways Clay Member. The uppermost part of the Kellaways Sand Member consists of pale greenish grey, fine grained sands and sandy silts, which are silty and clayey in parts. The beds are laminated and locally cross-stratified; bedding has generally been destroyed by bioturbation. Some beds are weakly cemented whilst others are friable and porous. The lower part comprises interbedded silty mudstones, silty sands and sandstones, the coarser beds being indistinguishable from the beds above. The Kellaways Clay Member consists of medium to dark grey, smooth, silty mudstone with indefinite silt laminae and scattered small bivalves and gastropods. Ill-defined mottling and silt-lined burrows occur throughout. Compared with the lower Oxford Clay, the member is more uniform, non-bituminous and poorly fossiliferous.

The Cornbrash consists of fine-grained sparry limestones (c. 80 per cent) interbedded with thin marls (c. 20 per cent). The limestones, which are hard, compact and crystalline, contain abundant shell debris and scattered shells in a microcrystalline matrix with micrite pockets. The marls consist of thinly bedded, dark grey medium to fine-grained shell-debris with crushed bivalves set in a clayey matrix with clay partings. They are generally friable or very weakly cemented.

The upper part of the Forest Marble consists of pale greenish grey, smooth mudstones with layers of fine silt, disseminated mica, plant debris, brown chitinous fish debris and, less commonly, shell-pavements of disarticulated bivalves. The number and thickness of the silt-debris partings varies and in places they coalesce to form lime-cemented ribs and lenticles. Increase in the lime content within the mudstones causes slightly more greyish hues and more blocky fractures. The lower part of the Forest Marble consists of grey, medium grained, shell detrital oolites with clay wisps and mud flakes. The junction between the mudstone and the limestone is sharp.

The White Limestone consists of a series of grey to light grey limestones which are generally oolitic or peloidal and contain considerable shell debris. There are numerous small vugs and shell cavities, some of which are calcite filled, a number of open fractures were observed and many calcite veins. The limestones are separated by thin mudstone or marl layers. The limestone sequence combining the lower part of the Forest Marble and the White Limestone can be considered as a single hydrogeological unit, which we refer to as the 'Great Oolite' limestone.

Figure 5 shows a simplified geological cross section through the Down Ampney Fault. The strata adjacent to the fault were proved, by both phases of drilling, to be essentially horizontal and the lithological units are of fairly uniform thickness. Comparison of sequences from boreholes well away from the fault have allowed the total displacement of the fault to be calculated as between 48-50m.

In total eight boreholes have been drilled through the fault zone. The upper part of fault zone is readily identified by strata of increased dips which start at 5-10° and increase to about 70°, and occasionally as high as 85°. However this increase is not progressive with depth, sections with low dip alternate with sections with higher dips, although there is a general trend of more steeply dipping strata at greater depth. The identification of all fault planes is hampered because of the similarity of much of the Oxford Clay lithology, the fault planes can only be positively identified where there is clear evidence of shearing or different lithologies are juxtaposed.This has prevented the correlation of particular fault planes between different boreholes. In general four or five fault or shear planes can be identified in each borehole and these tend to be located towards the base of the fault zone. Boreholes which pass into clearly distinctive strata below the fault zone, such as DA10 and DA19 into the Kellaways Beds and DA11 into the Forest Marble, show a very rapid transition into essentially horizontal undeformed strata, with less than a metre of inclined beds.

This structure suggests the fault zone can effectively be divided into two zones (Figure 5). The upper part of the fault zone which consists of disturbed dipping beds and probably small fault planes with short displacements, which fault similar lithologies against each other and are difficult to identify. The main fault zone consists of steeply dipping strata and contains the fault planes of larger displacements which clearly juxtapose different lithologies. The base of the main fault zone is essentially sharp, shearing against basically undisturbed strata below. Correlations between the borehole cores indicate the fault dips to the north at about 70°. The top of the fault zone was not clearly identified due to the lack of complete coring, but it seems likely that the disturbed zone has a vertical thickness of approximately 10m, which gives a zone thickness of about 3.6m. The main fault zone has a vertical thickness of between 3.8 and 11.5m with an average of 6.6m giving a zone thickness of approximately 2.4m.

The reconnaisance geophysics survey (Jackson et al, 1989) showed the strike of the fault to be east-west. In the absence of any specific evidence concerning the direction of displacement on the fault plane, it is assumed there was no strike slip movement and that the fault is consequently a normal fault.

6 Geophysical characterisation of the Down Ampney Fault

A detailed investigation was initiated into the possibility that the clays possessed a marked electrical resistivity anisotropy (Raines et al, 1989). This followed the observation of a discrepancy between resistivity depth sounding data and a model based on pre-existing borehole resistivity data and the bending of the Oxford clay beds in the fault zone.

The anisotropy of electrical resistivity of the Oxford Clay was examined by a series of measurements of resistivity on the core with current flowing horizontally and vertically. The horizontal resistivity core data was found to agree closely with the downhole resistivity logs. This enabled the implementation of a simplified methodology where vertical resistivity was measured directly on intact core immediately after extrusion from the core barrel, while the horizontal resistivity was measured from downhole resistivity logs.

Sections through the Oxford Clay in the faulted zone and to either side were investigated and it was found that the anisotropic resistivity ratio was constant either side of the fault at a value of 2-3 (Figure 6), but reduced to 1 or less in the fault zone. If the anisotropy is caused by reduced electrical resistance along the bedding planes then the observed change in ratios could be caused by the inclination of the bedding planes in the fault zone. The increase in horizontal resistivity in the fault zone may explain the resistivity peaks seen at the position of the fault in the earlier resistivity reconnaissance surveys. Furthermore this increase within the Oxford Clay could be detected directly by shallow surveys and used as a means of locating faults in thick sequences of clays.

Two short additional high resolution resitivity traverses were carried out perpendicular to the fault (Hallam et al, 1989), one of which was located along the line of the measurement array. In each case a constant electrical field was maintained with stationary current electrodes (AB=300m) with one potential electrode also held stationary, approximately in the centre of the array, the other was traversed to measure the relative total electrical resistance field. The general gradient or trend is then subtracted from the measurements to leave residuals. These residuals are shown for the whole of line N in Figure 7a and for the central portion in Figure 7c.

This technique enables the mean gradients for each side of the fault to be extrapolated to an intersection point, with a precision of better than 1m. This represents the most precise 'location' for the fault yet attained by surface geophysics. Between the two mean electrical field gradients there is an intermediate zone, which for line N falls between -4 and +1m (Figure 7c). The geological section constructed from the borehole array indicates that this zone lies above the intersection of the main fault planes with the Kellaways Beds. This spatial relationship must be regarded as empirical, given the complex structure of the fault and its consequent effects on the resistance field.

A similar field technique was used to make detailed two dimensional multiple resitivity measurements in an area 50m square which straddles the fault adjacent to the boreholes (Hallam et al, 1989). Two orthogonal pairs of 300m spaced AB electrodes were positioned parallel to and perpendicularly across the fault. A reference electrode was the placed in the centre of the two pairs. These five electrodes then remained fixed while measurements of potential difference from the central electrode were taken in the box at 2.5 m spacings along lines 5m apart perpendicular to the fault. Measurements were taken using first one pair and then the second pair of AB electrodes, thus giving measurements of potential differences with current flow parallel and perpendicular to the fault. The resultant resistivity values can then be contoured as in Figure 8 which clearly shows the fault. A subsidary feature was recognised in the fault which was investigated by rotating the direction of current flow by combining the two data sets and by directional illumination to enhance diagonally orientated features. This suggests that the structure of the fault may be complex and may contain blocks of material with significantly different physical properties.

Cross-hole seismic tomography surveys have been completed in the borehole array at Down Ampney (Jackson et al, 1989b) with the twin aims of imaging the fault plane to examine its geological structure, and determining changes in physical properties of the clay caused by the faulting. The measurement borehole array was designed to allow both the gross lithological changes across the fault, and the clay fault zone itself to be investigated by the cross-hole seismics.

The Oxford Clay was expected to have the lowest velocity, being in the range 1.6 - 1.7 km/s with the Kellaways sands approximately 10% higher. The underlying Cornbrash limestone was thought to have a far higher velocity in the range 2.5 - 3.0 km/s. Thus the large velocity contrast between the fault zone and the Cornbrash is likely to mask subtle details of the fault structure. However where the fault zone lies between the Kellaways and the Oxford Clay subtle changes associated with alteration of clay properties may be detectable.

The environment at Down Ampney was favourable for cross-borehole transmission seismic tomography, with no man-made background noise, and very low attenuation of the seismic waves. A high resolution borehole seismic source and a multi element hydrophone array was used for the surveys. This propagated energy of 1.5m wavelength in the Oxford Clay, representing a frequency of 1.2KHz which is far higher than that used in standard site investigation practice. The noise levels were low enough to enable whole waveforms to be acquired which often exhibited both reflections and refractions from the thin Cornbrash layer, indicating the benefits of underground seismic surveys that are not degraded by thin attenuating surface layers.

The effects on the velocity re-construction of using different arrivals were studied, and the removal of those events which were critically refracted along the Cornbrash improved the velocity inversion. Even though the geological setting was very favourable, it was concluded that the simplest straight ray interpretation procedure produced the best results with the real field data. The finite wavelength and the uncertainty in the exact position and shape of the refracting zones affected by the fault reduced the benefits of bent ray modelling. This is an unusual situation as the majority of reported case histories show that bent ray modelling produces results that identify geological structures more clearly.

The results from Down Ampney show a zone of 5% lower velocity within the clay coincident with the fault zone observed by the geological logging of the core. This lower velocity suggests that the physical properties of the clay within the fault zone are altered, having a higher porosity and lower strength. This implies that the hydraulic conductivity of the fault zone could be greater than the surrounding clay. Furthermore the reconnaissance resistivity surveys showed fault responses thought to be due to altered zones within the clay, that were of far greater magnitude than at the site of the borehole array. This suggests that the fault zone detected by the seismic tomography may be atypical of the Down Ampney site on a larger scale, and that the changes in physical properties may be greater in other parts of the fault.

7 Hydrogeological characterisation of the Down Ampney Fault

The geological strata in the Down Ampney region dip gently towards the SSE. The outcrops of the Great Oolite limestones and the Cornbrash form hills to the north and the Corallian outcrop forms hills to the south. Figure 9 shows a schematic section and the anticipated directions of groundwater flows. Down Ampney is situated at almost the lowest point of the Thames Valley where it would be expected that groundwater flow was upward, driven by the higher potentials of the recharge areas to the north, and that the Cornbrash and Great Oolite aquifer units were artesian.

Groundwater heads and hydraulic conductivities have been measured at Down Ampney (Brightman et al, 1989b) to investigate the local groundwater flow regime. The array of measurement boreholes (Figure 4) was designed to provide three vertical profiles of groundwater head. One profile was constructed through the upthrow side from the two piezometers, DA12 and DA20, in the overlying clay and the boreholes, DA10, DA11 and DA18, with screened sections through the Cornbrash and Great Oolite. The second was constructed through the downthrow side from the nest of clay piezometers, DA13, DA14 and DA16, and borehole DA15 with a screen through the Cornbrash. The third was constructed through the fault zone itself from the four piezometers, DA22, DA19, DA21 and DA17.

The groundwater levels measured in the boreholes with screened sections through the Great Oolite and the Cornbrash, on both sides of the fault, were well below ground level, see Table 1. These extremely low heads, compared to the artesian conditions which might have been expected, are caused by groundwater abstraction for public water supply by two pumping stations a few kilometres distant.

Upthrow side

Depth (m below gl)	Formation (m above gl)	Groundwater head conductivity	Hydraulic conductivity (ms^{-1})
14.1-16.2	Oxford Clay	-0.2	$9\mathrm{x}10^{-11}$
28.2-30.3	Kellaways	1.7	$9\mathrm{x}10^{-12}$
32.1-37.0	Cornbrash	-6.0	$1\mathrm{x}10^{-5}$
48.9-85.0	Great Oolite	-11.0	$5\mathrm{x}10^{-5}$

Fault zone

Depth (m below gl)	Formation	Groundwater head (m above gl)	Hydraulic conductivity (ms^{-1})
14.3-16.3	Fault zone	4.9	---
22.7-25.2	Fault zone	3.0	$2\mathrm{x}10^{-10}$
28.8-30.8	Fault zone	0.8	$2\mathrm{x}10^{-8}$
42.2-44.9	Fault zone	-1.53	$4\mathrm{x}10^{-11}$

Downthrow side

Depth (m below gl)	Formation	Groundwater head (m above gl)	Hydraulic conductivity (ms^{-1})
23.0-25.1	Oxford Clay	3.5	$4\mathrm{x}10^{-12}$
49.0-51.4	Oxford Clay	5.2	$6\mathrm{x}10^{-12}$
67.6-69.9	Kellaways	2.4	$3\mathrm{x}10^{-11}$
82.8-88.1	Cornbrash	-10.0	$4\mathrm{x}10^{-6}$

Table 1 Groundwater heads and hydraulic conductivities measured at the Down Ampney Fault Research Site.

Groundwater heads were measured in the piezometers by two methods. Where groundwater overflowed the top of the piezometer the tube was sealed by fitting a valve and the head measured using a pressure transducer. Where the groundwater level was below ground level, a transducer with an integral mini packer was lowered below the water level and the packer inflated by compressed air. Both these methods produce a sealed measurement interval which has a low compressibility, and consequently very little groundwater needed to flow in or out of the the low hydraulic conductivity clay test sections to reach an equilibrium head condition. Even so the zones were monitored for 4 to 5 days to obtain reliably stable head measurements. Figure 10 shows the trends of groundwater head which were measured. At first sight these do not appear to support either upward or downward groundwater flow. The most likely explanation seemed to be that the groundwater heads in the low hydraulic conductivity clay had not reached equilibrium with the heads in the aquifer which were generated by relatively recent pumping effects. In this interpretation the heads distant from the aquifers on either side of the fault (DA12 & DA20 and DA16 & DA13) were relics of the natural groundwater flow system and showed upward groundwater flow. If the hydraulic conductivity of the fault zone is enhanced then the groundwater head will re-attain equilibrium with the aquifers more quickly, which is consistent with the observed trend of heads decreasing with depth. The deepest piezometer on the downthrow side, DA14, is extremely close to the fault zone and may also have begun to re-equilibrate as a consequence.

This hypothesis to explain the groundwater head distribution was tested by mathematical modelling (Sen, 1989) using values of the hydraulic conductivities and storativities measured at the site. Hydrogeological parameters and groundwater level data further away from the site was much less detailed and insufficient to assist the modelling precisely. The aim of the modelling was to see if the measured head distribution trends could have been generated by pumping the Great Oolite, rather than to attempt to reproduce it precisely.

A 2–D vertical section about 2km long and 100m deep was modelled using the finite element code FEMWATER. On the upthrow side the geology was modelled as a sequence of horizontal strata down to the Lower Fullers Earth/Taynton stone boundary (Figure 5). The downthrow side was similarly modelled down to the Forest Marble/White Limestone boundary at practically the same depth. The two sides were separated by a slanting line of elements representing the fault. The clay and other impermeable layers were assigned conductivities in the range 10^{-10}–10^{-11} ms^{-1} and the limestone layers 10^{-5}–10^{-6} ms^{-1}. The fault elements were given various properties in order to simulate the measured head pattern. The top surface at ground level was given a fixed head boundary condition of 0m and the lower surface was taken to be a no–flow boundary. The boundary conditions at each end of the model were altered for various simulations.

Initially steady-state simulations were performed but these concluded that the positive head values measured in the clay could only result from unrealistically high heads imposed at the boundaries. Therefore transient simulations were undertaken to see if these could be remnant heads from before the present water-supply pumping regime.

The water abstraction history was modelled as a simple step drawdown from heads of +10m in the White Limestone to the present -11m around 30 years ago. This simplification was as accurate as was justified by the available data. A number of simulations showed that the measured heads were consistent with this model if the fault was assigned a low conductivity of approximately 10^{-10} ms^{-1} except between the downthrown Cornbrash and upthrown White Limestone where it was given an intermediate value of 10^{-8} ms^{-1}. In addition it could be observed that the head gradients in the low permeability layers stayed vertical except at very small distances from the fault (less than the thicknesses of the layers themselves) and that even a narrow fault zone of low or intermediate permeability can cause a significant head drop across it.

Hydraulic conductivity was measured in the aquifer units by constant rate abstraction pumping tests and in the clays by pulse tests (Brightman et al, 1919b). The pulse tests were performed in the overflowing piezometers by simply opening and closing the valve at the surface, and by measuring the volume of water released the compressibility or effective radius of the system can be measured. The pulse test were performed in th packered piezometers by rapidly deflating and inflating the packer to compress the water in the test section. By filling the piezometer with water before the test the compressibilty can be calculated by the decrease in water level, assuming the packer inflates to the same volume. The results show that the hydraulic conductivity of the fault zone tends to be between one and two orders of magnitude greater than the adjacent unfaulted clay, Table 1.

Pore water has been squeezed from clay core samples and groundwater samples have been collected during pumping tests in the large diameter boreholes and from piezometers. These samples have been analysed for major elements and stable isotope compositions (Ross, 1989) and profiles of variations in pore water and groundwater chemistry on both sides of the fault and through the fault itself have been constructed. These profiles suggest that the fault has had a significant effect on solute transport.

8 Summary and conclusions

The main conclusions that can be drawn from the Down Ampney fault study are :-

1. Electrical resistivity surveys have proved able to identify and map the location of major faults in clays.

2. Electrical resistivity surveys are unable to provide detailed information about specific faults, such as precise lithological thicknesses, the exact fault location and fault dip. This information is vital to the careful planning of more detailed hydrogeological and geophysical investigations and is best obtained by drilling cored boreholes.

3. The clays at Down Ampney have a resistivity anisotropy ratio of between two and three.

4. Seismic tomographic imaging of the fault shows the fault zone has altered physical properties.

5. The physical characteristics of the Down Ampney fault are :-

    i) It is an east-west striking normal fault with a downthrow of c. 50m to the north.
    ii) The fault plane/zone dips about 70° to the north.
    iii) There is a zone of disturbed and slightly fractured rock of c. 5-10m thickness.
    iv) The main fault zone is heavily sheared and fractured and is c. 1-2m thick.

6. The groundwater flow system at Down Ampney is not in a steady state. The groundwater heads in the clays are not in equilibrium with the heads in the aquifers.

7. The hydraulic conductivity of the fault zone is increased above that of the surrounding clay by between one and two orders of magnitude.

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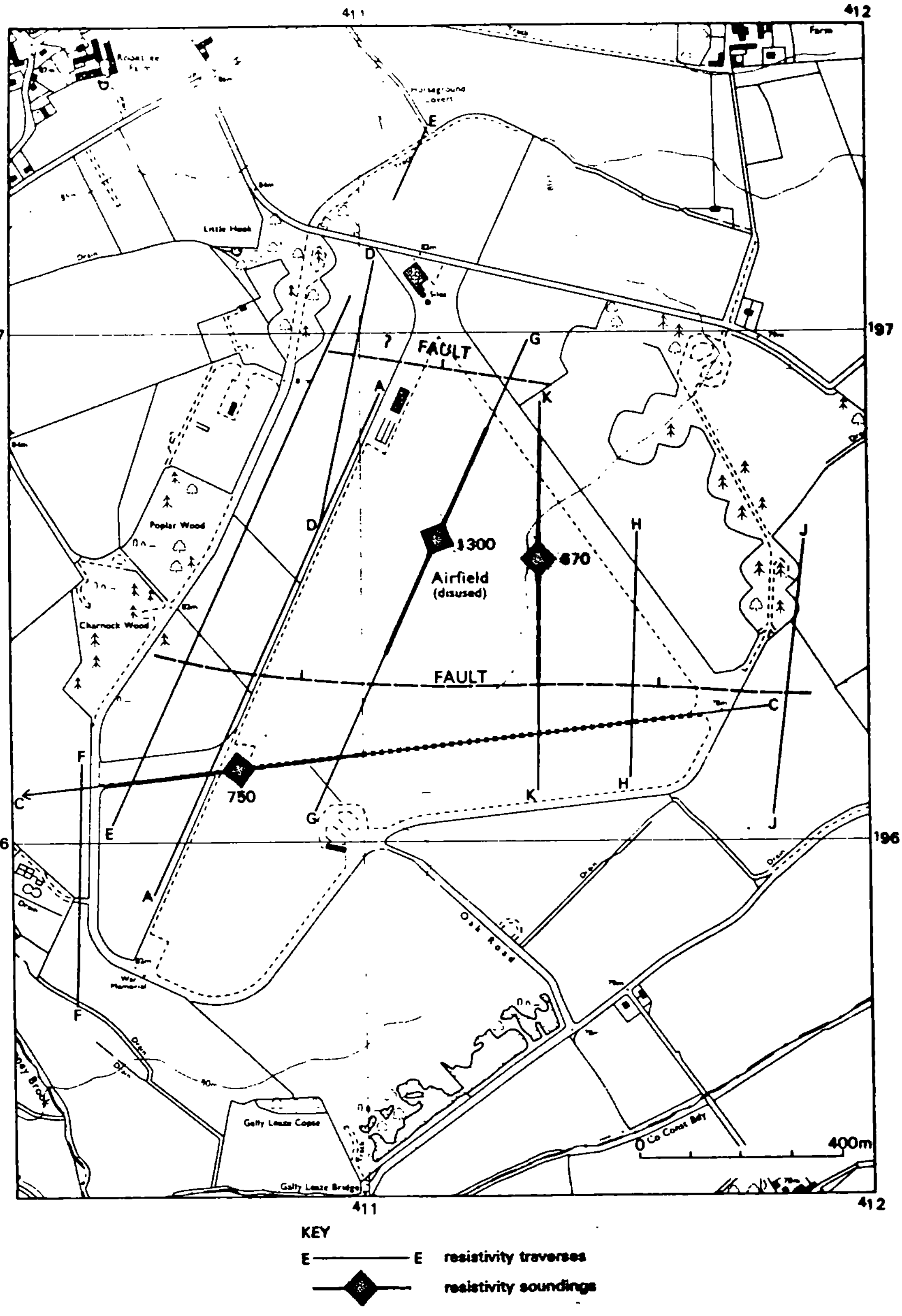

Figure 1 Location map of the resistivity traverses at Down Ampney.

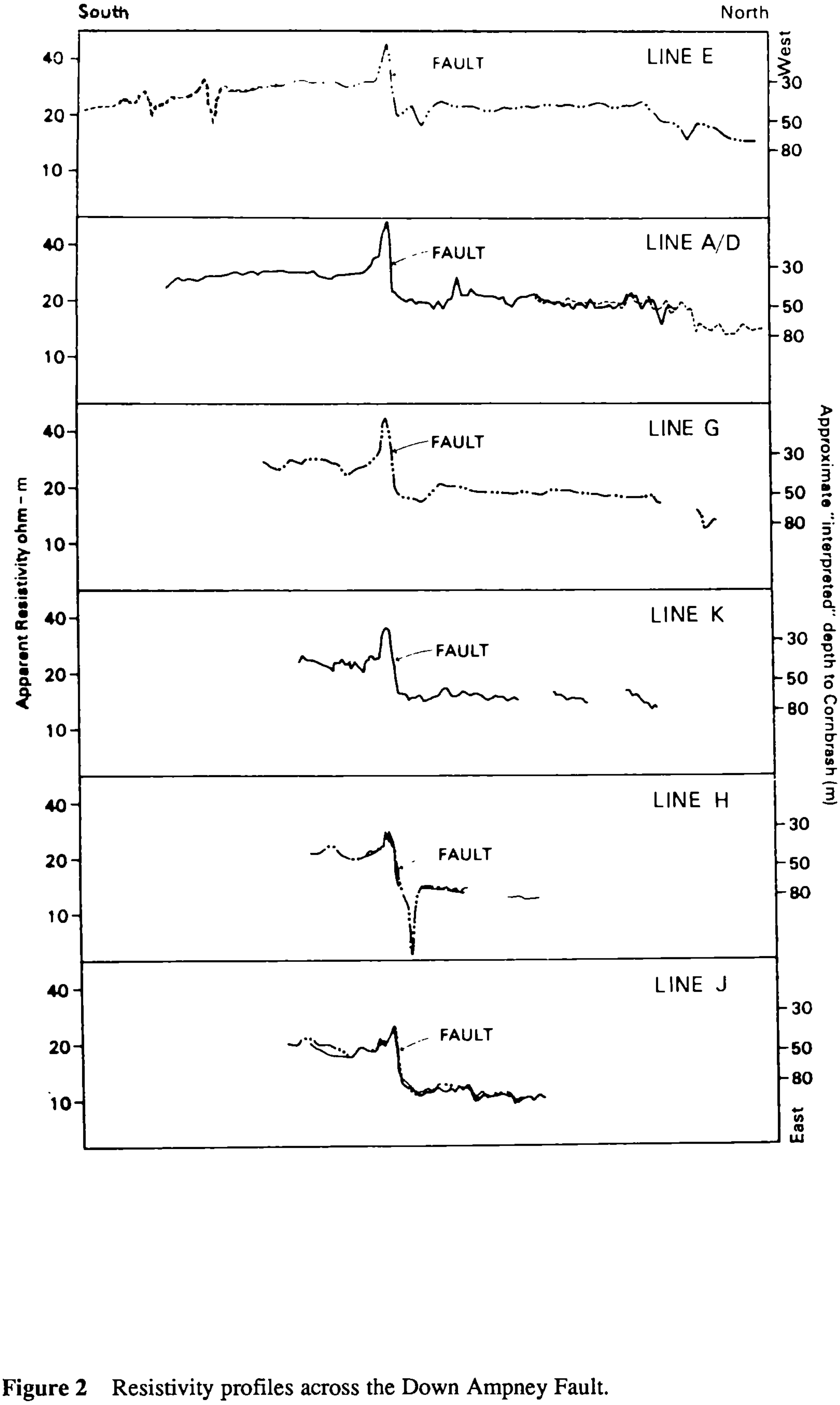

Figure 2 Resistivity profiles across the Down Ampney Fault.

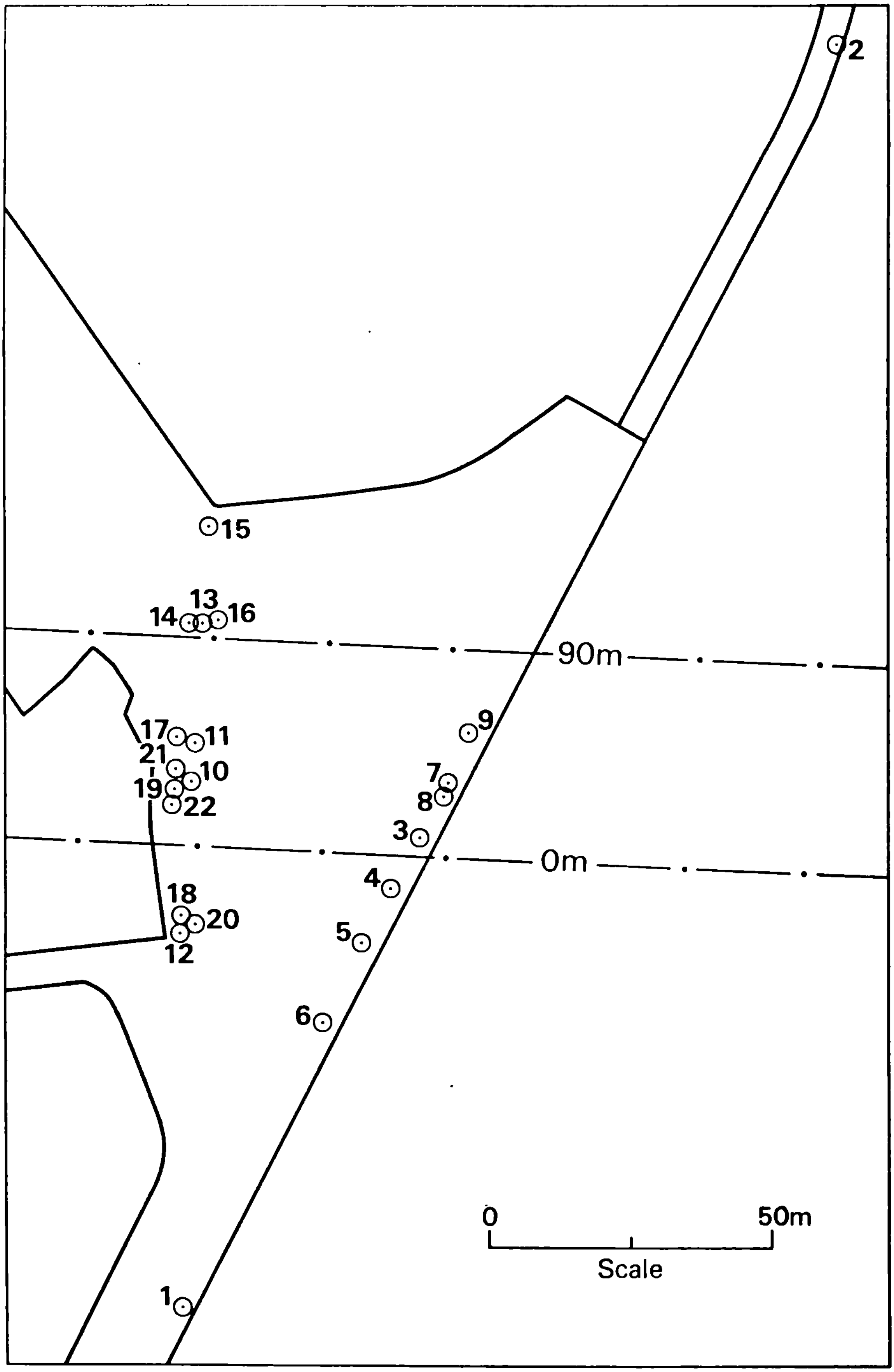

Figure 3 Location plan of the boreholes at the Down Ampney Fault Research Site.

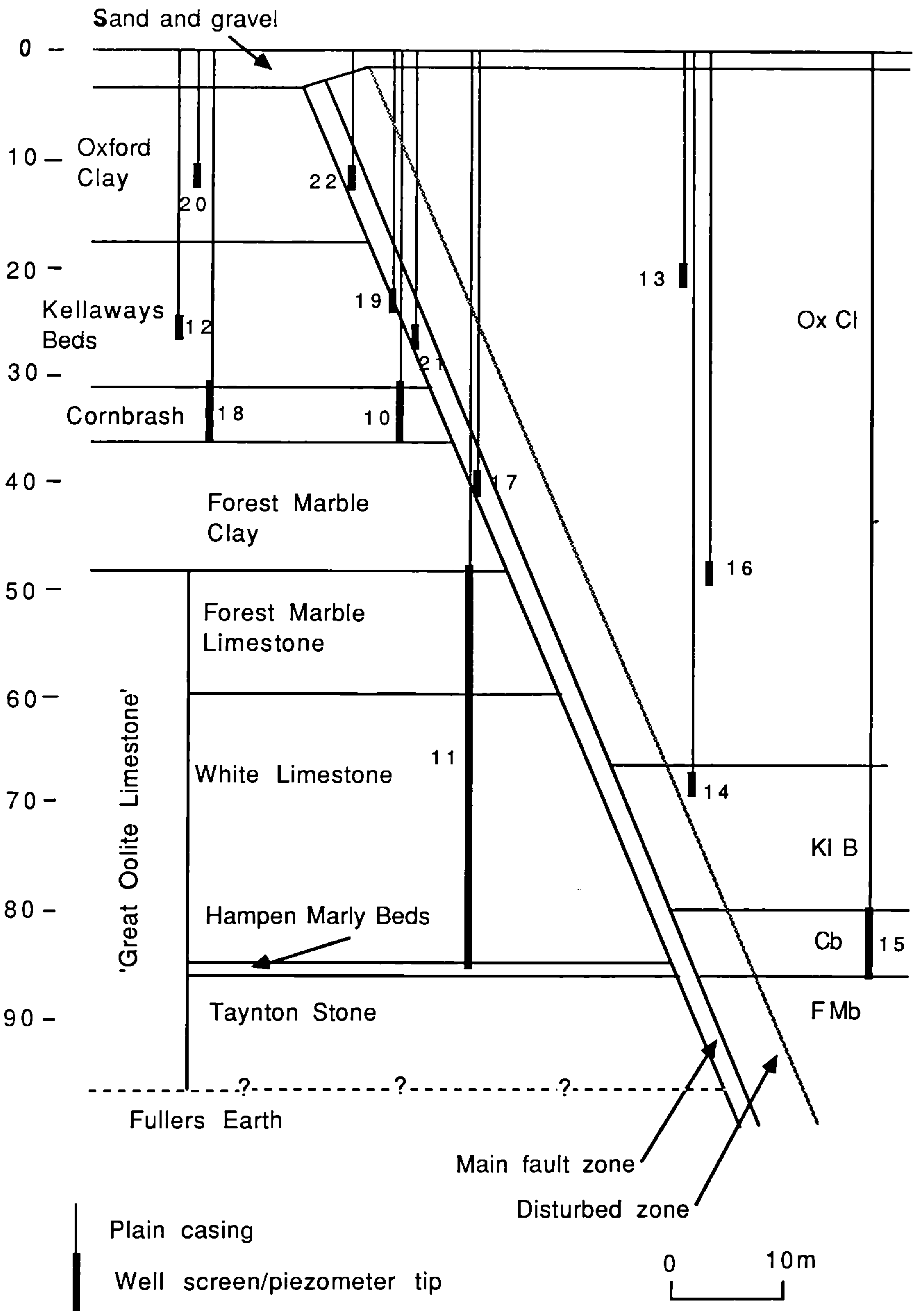

Figure 4 Schematic section through the measurement borehole array at Down Ampney.

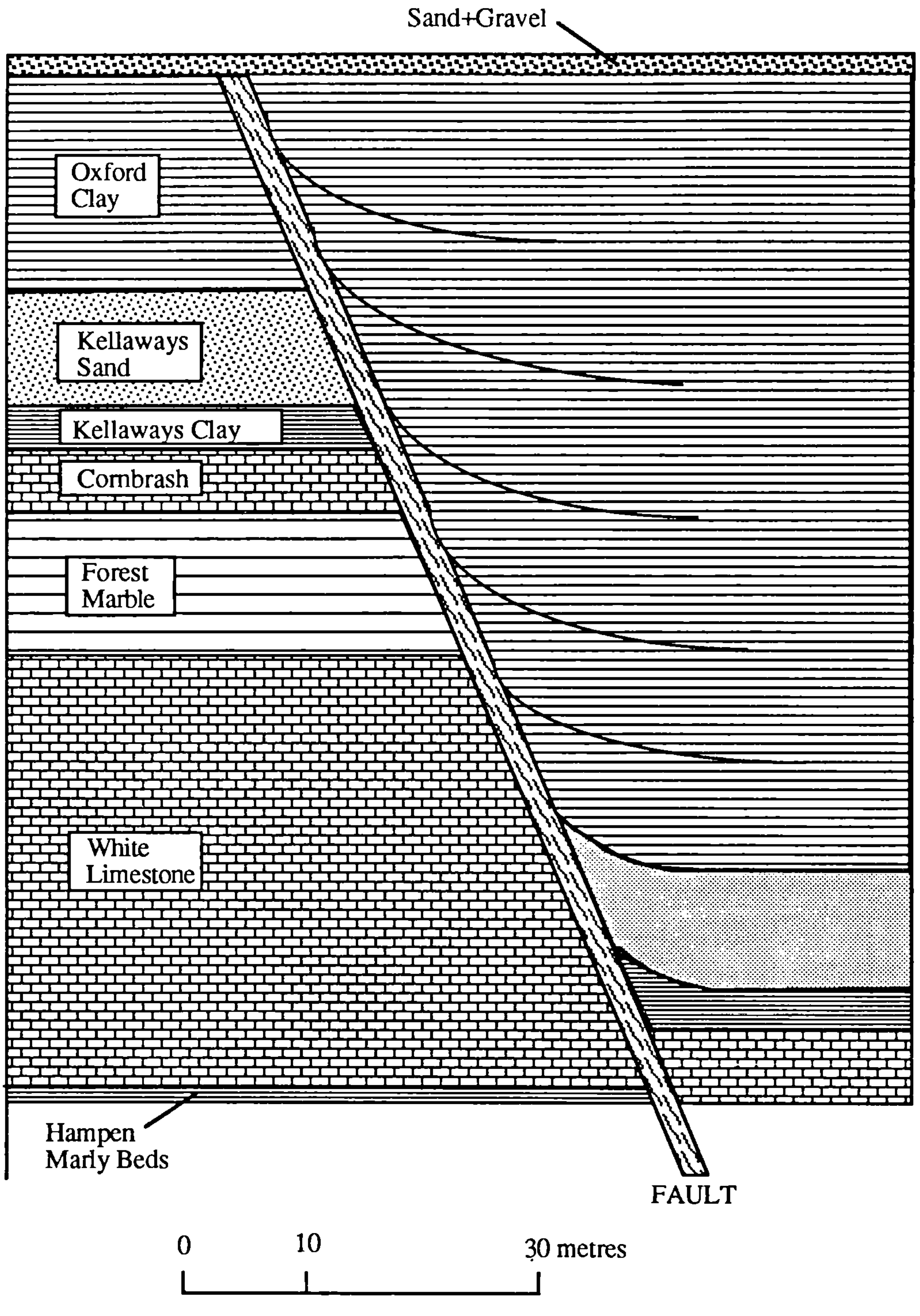

Figure 5 Schematic geological section through the Down Ampney Fault.

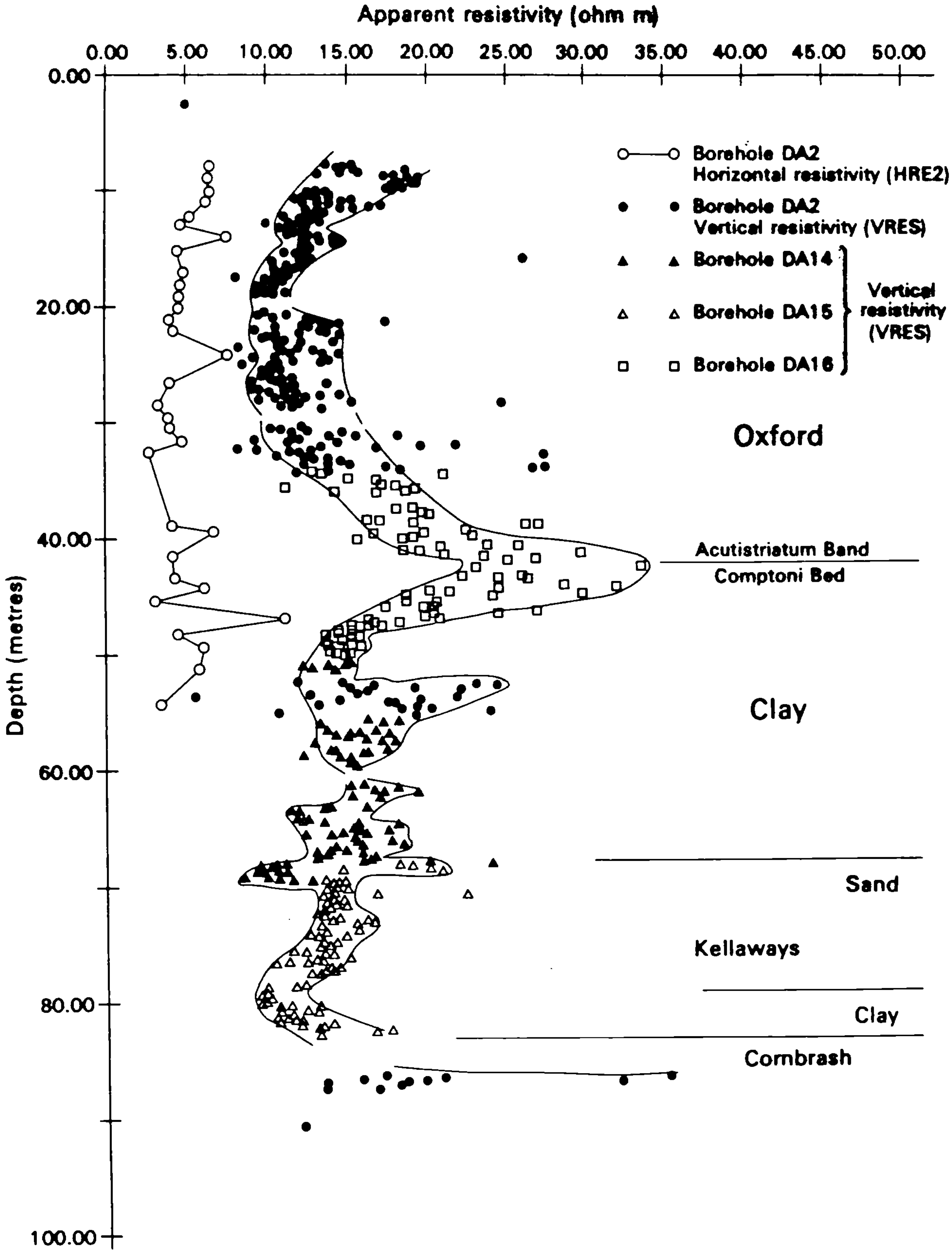

Figure 6 Composite of the measured vertical and horizontal core resistivities on the downthrow side of the fault.

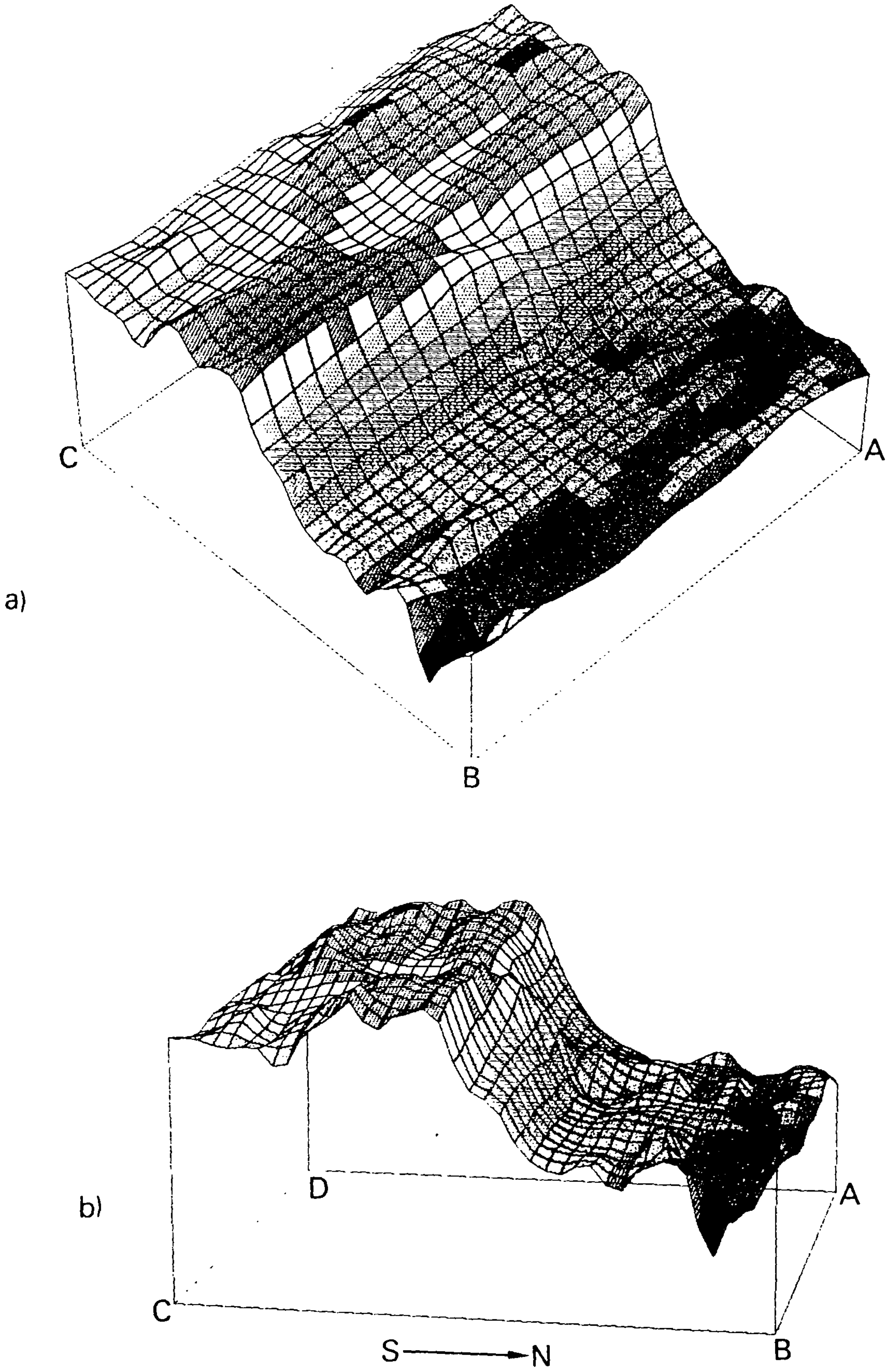

Figure 7 The Total Field Resistance measurements along line N at different spatial scales, after the removal of 0.3 ohm/m.

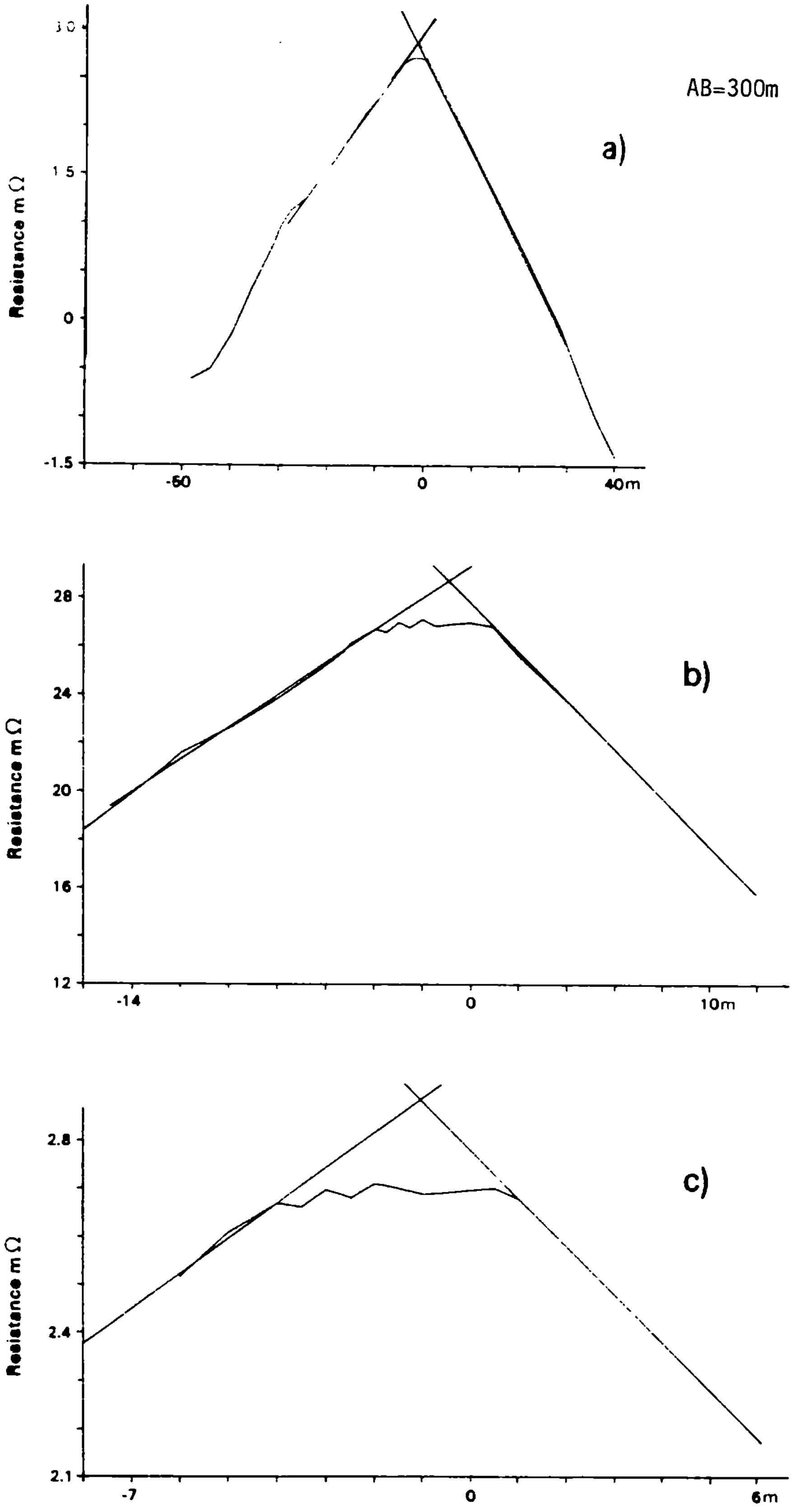

Figure 8 Box resistivity measurements with current flow perpendicular to the fault.

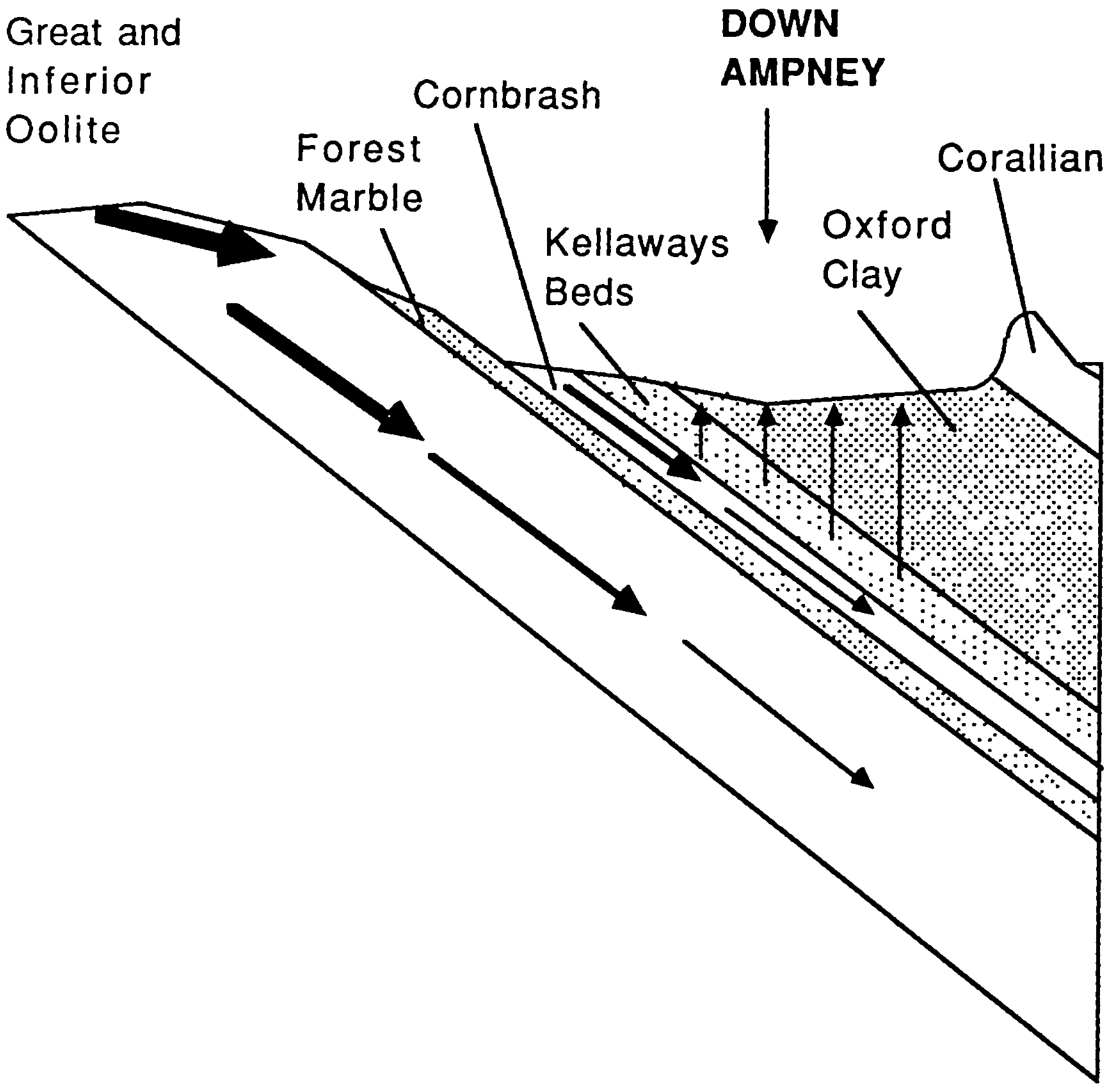

Figure 9 Schematic diagram of predicted regional groundwater flows.

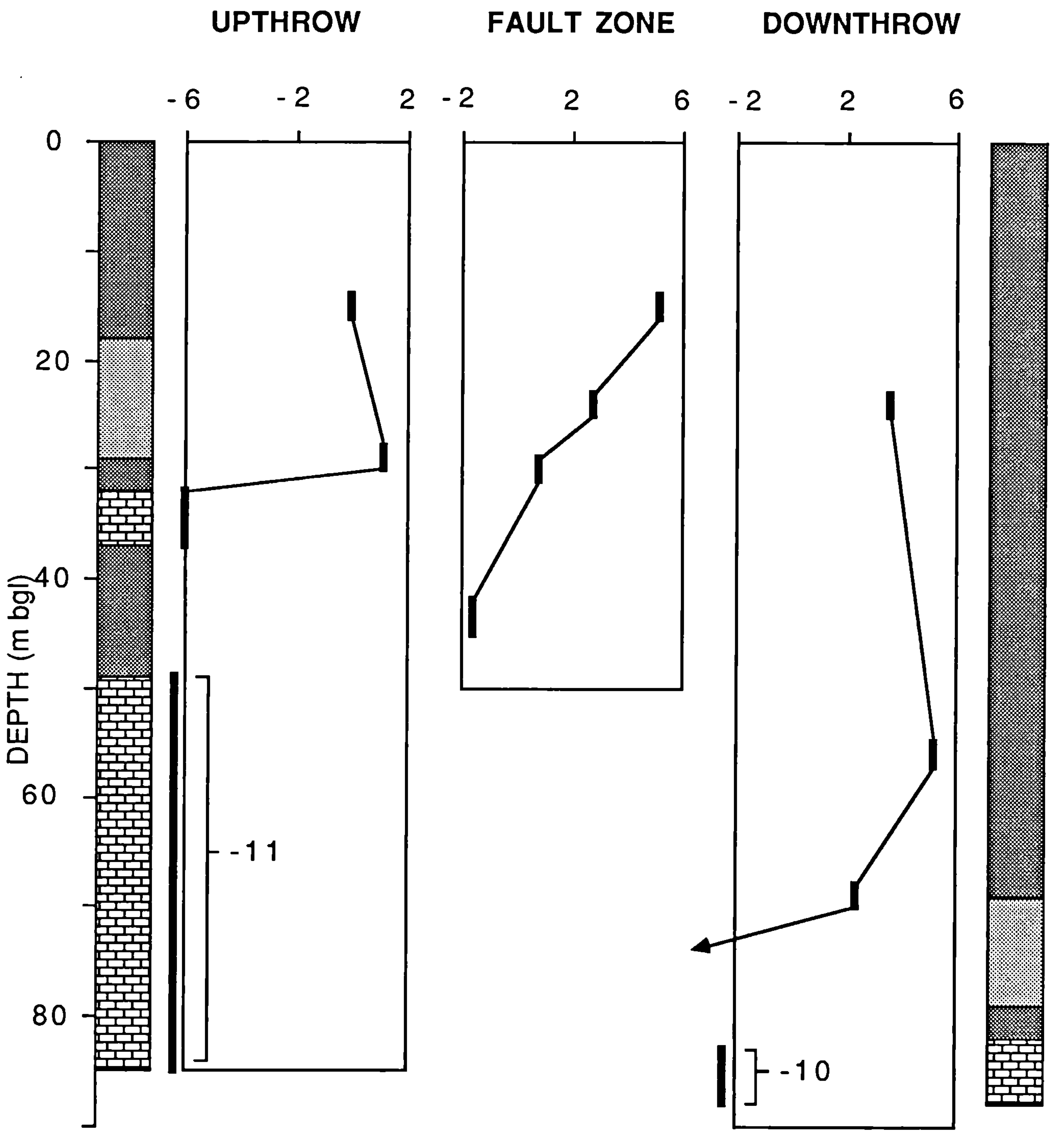

Figure 10 Groundwater heads measurements at the Down Ampney Fault Research Site.

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