Broubster (Scotland)

Description: There are a number of uraniferous localities in the Caithness area of northeast Scotland (Bowie et al., 1970; Gallagher et al., 1971), and the one at Broubster has received considerable attention from the British Geological Survey (BGS) as a natural analogue site for the migration of uranium radionuclides (Ball and Milodowski 1989; Higgo et al., 1989; Milodowski et al., 1989; Longworth et al., 1989; Read and Hooker, 1992; Read et al., 1993). Unfortunately, the site was destroyed by forestry activities in the late 1990s.

The uranium, lead and zinc mineralization at Broubster occurs 6 km southeast of the Dounreay nuclear site in a low-relief, marshy landscape of brown earths, peat and boulder clay soils (National Grid Reference ND 021 624). The land in the Broubster area is drained by surface and near-surface waters that flow gently eastwards into the Forss Water river, which discharges onto the coast at Crosskirk 7 km away to the north (Figure 1).

The solid geology at Broubster features rocks belonging to the Lybster Subgroup of the Lower Caithness Flagstone Group of the Middle Old Red Sandstone. The Caithness Flags are lacustrine sedimentary rocks comprising mudstones, siltstones, sandstones and limestones with low-angle northward dips. They are the result of non-marine sediments deposited about 400 Ma ago at the western margin of a large continental lake (Mykura, 1983).

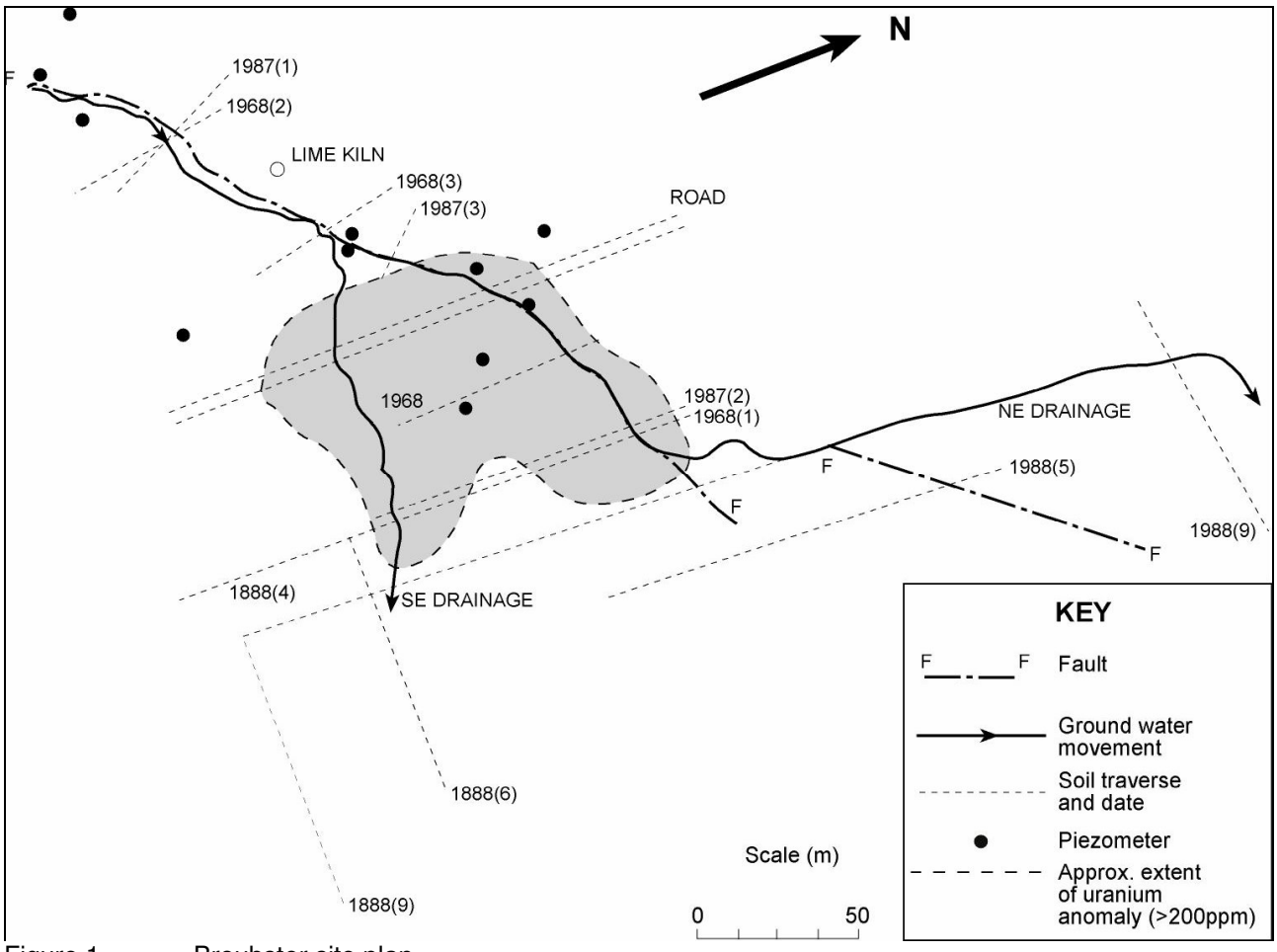

Figure 1 Broubster site plan

The detailed geology, hydrogeology and geochemistry of the Broubster site are described in Ball and Milodowski (1989) and Milodowski et al. (1989). Radiometric and geochemical investigations by the BGS in the late 1960s revealed uranium (and Zn and Pb) mobilisation, transport and dispersion downslope from a source in the bedrock close to the surface on the west side of the site into a peat-bog overlying boulder clay to the east. The site map (Figure 1) shows a minor road running north-south that divides the site between the near-surface source rocks and brown earths

to the west and the peat soils to the east. Gallagher et al. (1971) considered the source to be at the site of an old limestone quarry (close to an old lime kiln and largely backfilled). Gallagher et al. (1971) reported the occurrence of fault-bounded vein mineralization with a northeast trend hosted by a 0.4 m thick bed of bituminous limestone with similar strike and gentle dip to the north. Although the mineralization was found to comprise calcite with dolomite, sphalerite, galena and globules of uraniferous hydrocarbon, it was suggested that the bulk of the uranium and other metals in the secondary concentration in the peat were probably derived from the limestone itself.

Further BGS fieldwork from 1986 to 1991 delineated a northeast-trending fault that cuts the limestone bringing up massive calcareous sandstone with over 100 mg/kg of U. The U resided within apatite and dispersed U-Si-Ti phases often associated with pyrite and organic lamellae. Weathering had caused decalcification of the sandstone and leaching of metals from these source minerals. Uranium migration via groundwater transport and dispersion had occurred over a distance of over 100 m into the peat marsh (Figure 2). Read et al. (1993) thought that the bulk of the U had been transported as soluble U(VI) fulvic acid complexes (Smith et al., 1990). Once in the peat, the carboxylic acid functional groups on the humic substances successfully competed for the metal to produce a young uranium anomaly with over 0.1 wt % in some sections. The uranium distribution ratio between the peat and groundwater was measured to be c.10 m^3/kg (Higgo et al., 1989).

The secondary dispersion pattern closely followed the local drainage pattern of the site with an elongation of the peat U anomaly following a surface drainage channel flowing to the northeast. Ball & Milodowski (1989) also considered that, in the northeast of the site area where boulder clay cover was absent along the line of the fault, there was the potential for possible recharge of 'fresh' uranium into the peat horizon, either directly from bedrock (mineralized or otherwise) via the fault or from groundwater flowing out from the boulder clay-bedrock interface.

The history of the site before the last glacial deposition of boulder clay is uncertain, but there was probably a phase of glacial planing to produce a relatively flat surface. During the subsequent warmer climatic period from 10,000 to 5000 years BP this surface became forested. With the onset of cooler and wetter conditions, peat replaced the trees. A radiocarbon age of 4000 years BP was reported for the peat (Read et al., 1993). Secondary fixing of U in the peat probably began at this time and was accelerated considerably some 100-200 years ago with the quarrying of the limestone bed. Further liberation of uranium occurred in the 1970s and 1980s as a result of extensive liming of the local peaty soils west of the road. Liming would have raised the pH levels and released the uranium previously fixed in these soils (Ball & Milodowski, 1989). Finally, the whole site was radically changed in the 1990s when the whole area was ploughed up and drained for forestry planting. The final fate of the uranium in the peat bog was not investigated.

It took about 5000 years for the uranium anomaly to form in the peat bog at Broubster. The uranium mass accumulation in the peat occurred primarily through groundwater transport channelled largely through the fault zone. Assuming a 100 m zone length, a rough width of 2 m, a 20 m depth of source bedrock with an initial U concentration of 100 mg/kg and a rock density of 2.5 kg/l, then 1000 kg of U could have been leached and lost to groundwater if all the available U was leached out.

This calculation gives a time-averaged mass flux by aqueous flow from the bedrock of 0.2 kg U per year, equivalent to 2.5 x 10^6 Bq/yr of U-238.

If the uranium is dissolved to give a U groundwater concentration of 10^{-7} M (Read et al. 1993), which is equivalent to 0.0238 mg/l, then the volume of water required is 0.2 x 1E+6/0.0238 = 8.4E+06 l/yr.

Read et al. (1993) indicated that the Darcy velocity through the decalcified sandstone in the fault zone was 3 x 10^{-7} ms^{-1}. [Note that the unweathered bedrock was considered to be less permeable with a Darcy velocity of 4 x 10^{-10} ms^{-1}.] If a porosity of 0.3 is assumed for the weathered sandstone, then the linear groundwater velocity is 10^{-6} ms^{-1}, which is equivalent to 31 m/yr. The volume per

year that flows through the 200 m^2 fault zone is therefore 6200 m^3 equivalent to 6.2 x 10^6 l/yr, similar to the time-averaged estimate of 8.4 x 10^6 l/yr.

Using a mean uranium concentration of 400 mg/kg in the peat, an area of 2000 m^2 for the peat marsh, a density of 1.5 kg/l and a mean depth 0.5 m would give an estimate of 600 kg for the mass of accumulated uranium. This value is smaller than the bedrock source estimate of 1000 kg, probably as a result of U losses caused by the agricultural liming.

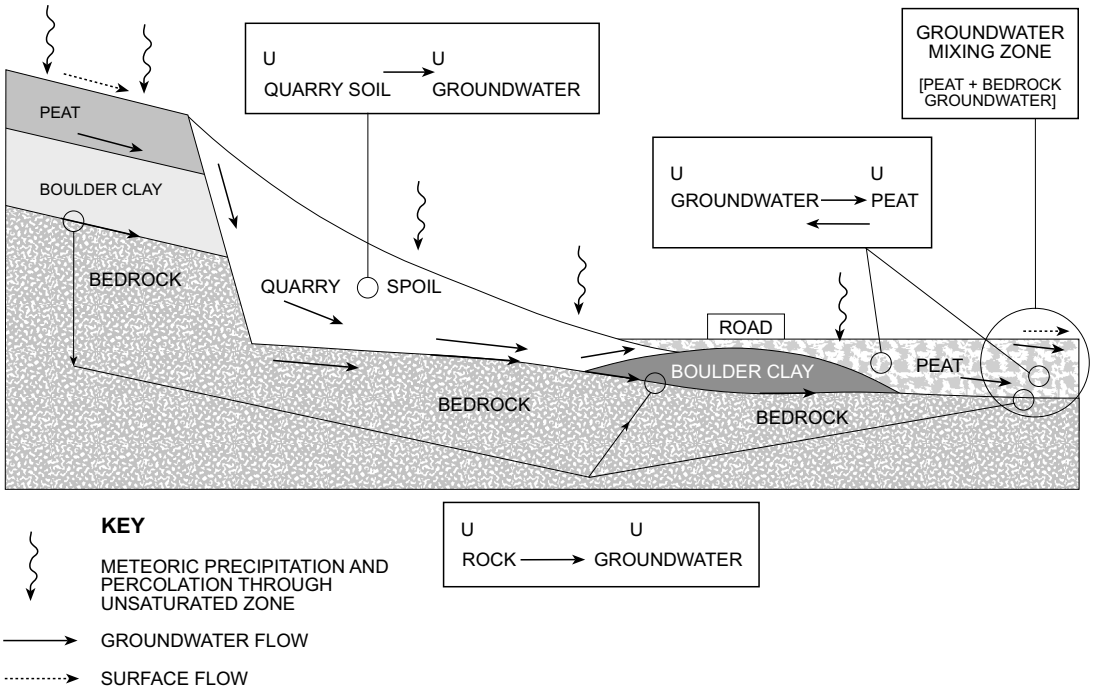

Figure 2 Schematic cross-section parallel to the fault

Relevance: The Broubster study is relevant for understanding the processes of near-surface migration of U by groundwater flow and its fixation in a peat bog in northern Scotland. Fulvic acid colloids were thought to play a major role in the aqueous transport of the U. The data were used to test speciation models for U.

The Broubster site is an example of a geosphere-biosphere interface (GBI), involving peat.

Position(s) in the matrix tables: The study occupies a box in the Geosphere matrix table.

Limitations: The study is limited to a near-surface/surface environment for U migration processes in a northern Scotland climate. The redox conditions were oxidising.

Quantitative information: Good quality concentration (mass and radioactivity) data for the bedrock, groundwater and peat/soil compartments for U and Th nuclides have been published. The source term sandstone (unweathered and decalcified) and limestone were analysed for their radionuclide compositions (U-238, U-234, Th-230, and Ra-226; Th-232 and Th-228). The thorium concentrations were similar in the fresh and weathered material (*ca.* 35 Bq/kg for both Th-232 and Th-228), whereas the uranium levels were depleted in the weathered and decalcified bedrock (1400 Bq/kg of U-238 in unweathered sandstone; 332 Bq/kg in decalcified sandstone). These rocks were also measured for other trace elements (Zn, Pb, As, La, Ce, Nd) – see Read et al. (1993).

Groundwater data for U recharging into and discharging from the fault zone are given in Read et al. (1993). Activity data for U-238, U-234, Th-230, Th-232 and Th-228 in filtered peat water are given

in Longworth et al. (1989). Little of the thorium had been moved by groundwater, in contrast to uranium. Whereas most of the U was in solution, any aqueous Th was largely on particulates or in colloid phases (Longworth et al., 1989).

Elemental concentration and total organic carbon (TOC) data for peat water are given in Ball & Milodowski (1989) and Read et al. (1993). Elemental concentration and TOC data for the fault recharge and discharge water data are listed in Read et al. (1993). Eh values varied from 175 to 386 mV.

The site had brown earth soils to the west of the road in the old kiln/quarry area and peat sediments to the east in the marshy area. The radionuclide activities of U-238, U-234, Th-230, Ra-226, Th-232 and Th-228 for a brown earth soil profile are reported by Read et al. (1993).

The measured radionuclide activities of U-238, U-234, Th-230, Th-232 and Th-228 in peat and boulder clay are reported in Longworth et al. (1989). These data were derived on samples collected from a profile in Pit 8 east of the road. The boulder clay data (B/C horizon at 39 to 49 cm below ground surface) indicate background values.

The U distribution ratio between the peat and groundwater was found to be *ca*. 10 m^3/kg by Higgo et al. (1989).

Uncertainties: There are some uncertainties associated with the past flow rates and compositions of the groundwater, especially in the fault zone. The disturbance of the geochemical system by liming of the land west of the road implies that the measurements of pH taken during the BGS sampling campaign from 1986 to 1991 did not reflect the lower pH values that had probably prevailed earlier during the formation of the uranium anomaly in the peat.

Time-scale: The time-scale of this natural analogue is geological i.e. Quaternary (specifically, Holocene <10,000 years). In particular, the processes of peat accumulation and uranium flux have occurred over the last *ca*. 5000 years.

PA/safety case applications: Previous uses of the analogue study in a PA or safety case may be included in reports by Nirex and BNFL. The UKAEA have considered the Broubster rock, soil and groundwater data in terms of background information for developing natural safety indicators in support of a safety assessment of a hypothetical LLW repository at Dounreay.

Communication applications: Previous uses of the analogue study in communication and dialogue material for different audiences are not known.

References:

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Added value comments: PA use of the Broubster site data in the development of natural safety indicators could be pursued further. PA use of the study as an analogue of U nuclide migration across a GBI into peat could be considered.

Potential follow-up work: Follow-up work in the field is no longer possible.

Keywords: Far-field, geosphere-biosphere interface, dispersion, colloids, U sorption, peat

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