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The significance of terrestrial volatiles as pathfinders in uranium exploration: examples from south-west England

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Evaluation of soil gas anomalies associated with sites of U mineralisation in south-west England shows that while convincing ^{222}Rn anomalies are commonly observed, the pattern of ^4He variation is less predictable. In addition, where ^4He anomalies do occur, they are only of comparable magnitude to those over metalliferous (but non-uraniferous) veins, or even non-mineralised fractures. Dissolved gas anomalies associated with U mineralisation are seen for both ^4He and ^{222}Rn . However, interpretation on anything greater than a local scale should be made in terms of structural and hydrological controls rather than mineralogical ones. The relationship between soil gases and dissolved gases is complex, and involves consideration of the solubility coefficients of all dissolved gas species. Exsolution to form discrete 'aerosol' phases may occur and cause the consequent scrubbing of trace gases with low solubilities. The degree to which this occurs is determined by the nature of the groundwater flow system.

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Introduction

The use of terrestrial volatiles in U exploration began with the successful application of ^{222}Rn activity measurements in the early 1960's, and recently reviewed by Gingrich (1984). Subsequently, many early research studies concerning ^4He were also in terms of the application of ^4He surveying to the identification of sites of U mineralisation (Dyck 1976; Reimer et al. 1979). A critical evaluation of ^4He surveys was also carried out by Butt and Gole (1984). The results of these later studies, which were far more comprehensive than the early research work, suggested that ^4He was an unsuitable pathfinder element in the application of U prospecting/surveying.

There are numerous occurrences of small-scale U mineralisation in south-west England, and the most well known localities have been documented by Rumbold (1954) and Dines (1956). However, more recent work by the Geological Survey has brought to light several new occurrences (Bowie et al. 1973).

When ^4He studies were first begun at Exeter, survey sites were selected with known or potential radiometric anomalies, in order to assess the use of the ^4He technique over areas of U mineralisation, and to compare this with other methods which are better understood. Comparative methods included soil gas ^{222}Rn activity measurements, dissolved U determinations and gamma-ray spectrometry. Five sites were investigated. Soil gas surveys for ^4He were carried out over the Fingle Bridge and Kingswood U deposits. ^4He concentrations in water as a dissolved gas were also carried out. The Fingle Bridge anomaly proved suitable for this method of investigation, and therefore allowed comparison with the soil gas method at the same survey site. High ^{222}Rn concentrations in springs in the Merrivale district of Dartmoor (SX550751) were also investigated, as were abstraction wells operated by the South West Water Authority at Taw Marsh (SX618906), which has a known high ^{222}Rn content, and in the Otter Valley, where abstraction takes place from a series of deep wells which sample groundwater from the Budleigh Salterton Pebble Beds and Otter Sandstone aquifer. Underlying these formations are the uraniumiferous nodule horizons of the Littleham Mudstone.

Survey results

At Fingle Bridge (Figs. 1 and 2), eleven soil gas sample sites with a 10m spacing were taken on a N-S traverse perpendicular to the most anomalous zone found by Geological Survey trenching (Fig. 2). The position of the E-W trending U veinlet is clearly shown by the total Rn ($^{222}\text{Rn} + ^{226}\text{Rn}$) activity data (14750 cpm, equivalent to 2950Bq1-1). Upslope of the vein, total Rn activities are minimal (13-67cpm, or 2.6-13.4Bq1-1). The ^{220}Rn activity at

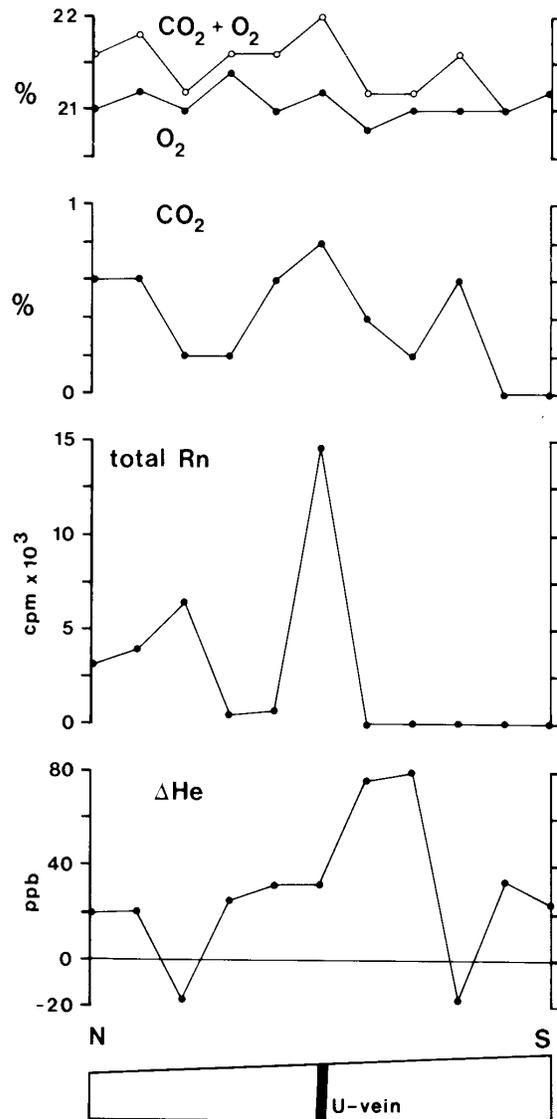


Figure 1. Soil gas variation on N-S traverse across U vein at Fingle Bridge.

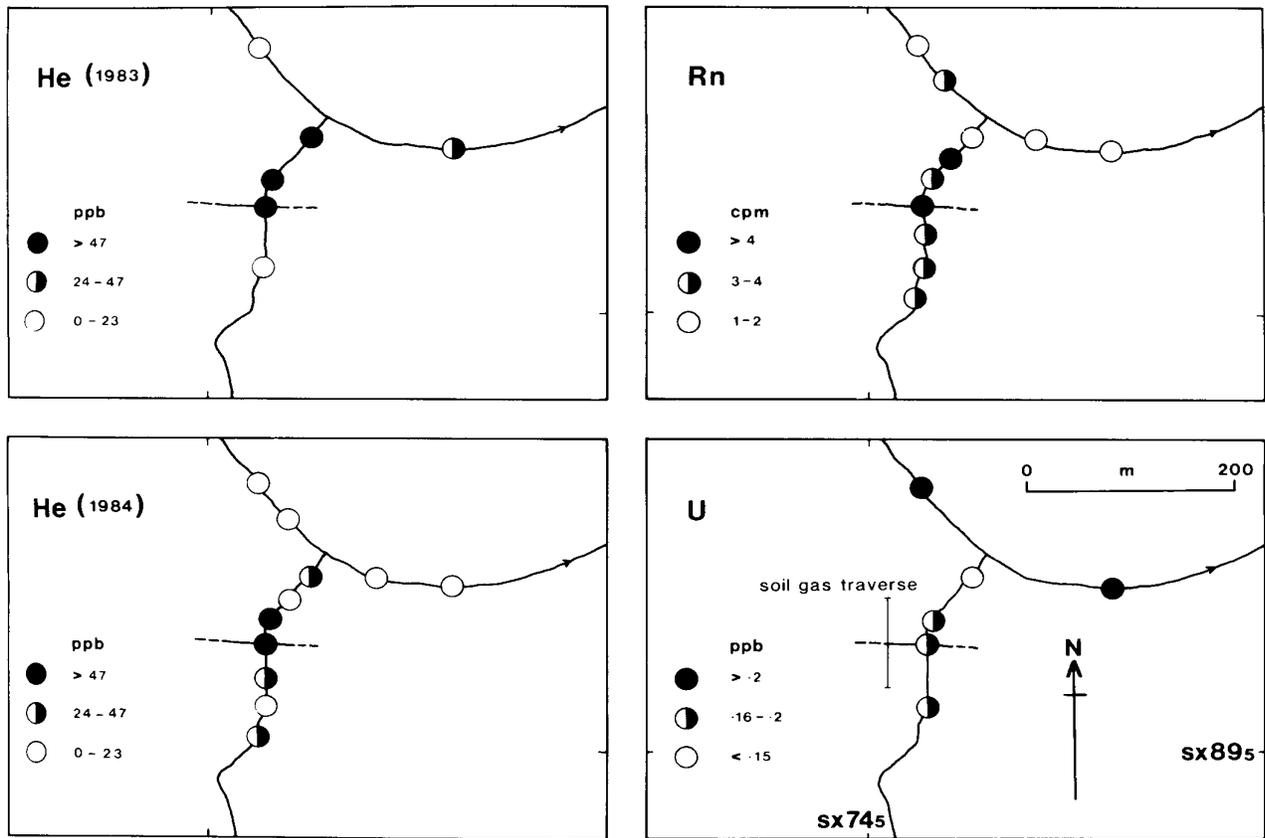


Figure 2. Dissolved helium, radon and uranium at Fingle Bridge. E-W trending vein shown solid where known, broken where inferred.

the Fingle Bridge survey site is negligible, and total Rn activity may be assumed to represent just ^{222}Rn . Downslope, some degree of overburden transport is suggested by the secondary peak, with activities between 415cpm and 6500cpm (83-1300Bq1-1).

Comparative variation in the other soil gases measured here show low variations, with ^4He ranging between -17 and +80ppb ΔHe (relative to the ambient atmospheric concentration). CO_2 and O_2 show small ranges in variation (0.0 - 0.8% CO_2 and 20.8 - 21.4% O_2). Only CO_2 shows a comparable peak associated with the sample site. This would be expected from typical soil gas behaviour, although the results from the other gases are equivocal. Correlation with gamma-ray spectrometry, however, supports the ^{222}Rn soil gas anomaly most effectively (Gregory 1987).

In addition to the soil gas survey, stream water samples were also taken for ^4He (on two occasions), ^{222}Rn and dissolved U analysis (Fig. 2). Here, ^4He in surface waters clearly shows the position of the vein where it crosses the stream draining into the River Teign. The significant value to note here is the 47ppb equilibrium concentration of ^4He in water. Values below 47ppb suggest turbulent degassing and/or dilution, as in the case of the mixing of stream and river waters. ^4He values range from 19-64ppb (July, 1983) and 0-60ppb (August, 1984). Corresponding ^{222}Rn activities are very low (1-7cpm, 0.3-2.1Bq1-1), but still delineate the position of the U vein. Dissolved U values are low for the stream (0.15-0.20ppb) draining Carboniferous sediments, but are higher in the main river (0.25ppb) draining the granite uplands.

At Kingswood, six traverses were sampled for ^4He . Two traverses (15 sample sites at 5m intervals) were over barren ground, to establish background variation in the area. This was in the range of 23-49ppb ΔHe (mean 37ppb, s.d. 9ppb). An additional four traverses (42 sample sites at 8m intervals) were run E-W across

the NNW-SSE trending vein (Fig. 3). Because of vegetational variations in the survey area, particular care was taken in the handling of results. The two most northerly traverses, over both woodland and scrub, show similar variation in the change of mean soil gas ^4He either side of the change in land use. The variation in mean values on all traverses suggests that each traverse should be taken on its own for assessment. While the most northerly traverse has the anomaly obscured by land use contrasts, the second most northerly of these has the expected peak over the vein. The third shows a significant peak, close to the predicted vein position, while the results on the fourth are equivocal. The site 'C7' marked on these two survey lines refers to one of the surface sample pits dug by Campbell (1947) and reported to have an above average radioactivity. This is not clearly shown on the ^4He data.

At Merrivale, comparison between ^4He in soil gas and associated groundwater proved possible around four high ^{222}Rn activity springs (Fig. 4). Soil gas ^4He was generally high (30-135ppb ΔHe), and all but one dissolved ^4He measurement was above the equilibrium solubility for ^4He (36-78ppb ΔHe). The Pearson correlation for these four sites, however, is only 0.185. Comparable ^{222}Rn variation from the springs is 85-392cpm (25.5117.6Bq1-1). This set of data correlate with dissolved ^4He at only 0.219, but with the soil gas at 0.911. These high ^{222}Rn activities are rapidly dissipated in the stream water, which shows ^{222}Rn activities of 2-38cpm (0.6-11.4Bq1-1). Again, dissolved U variation is low, from the detection limit (0.05ppb) to 0.40ppb, and correlations are also low.

Poor correlations between ^4He and U are not always the case. At Taw Marsh, three well water samples were analysed (Gregory 1987). Dissolved U contents are high (1.50, 1.55 and 3.35ppb), with comparable ^4He variation (39, 32 and 54ppb). Nevertheless, the results of ^4He determinations in the Otter Valley (Gregory and

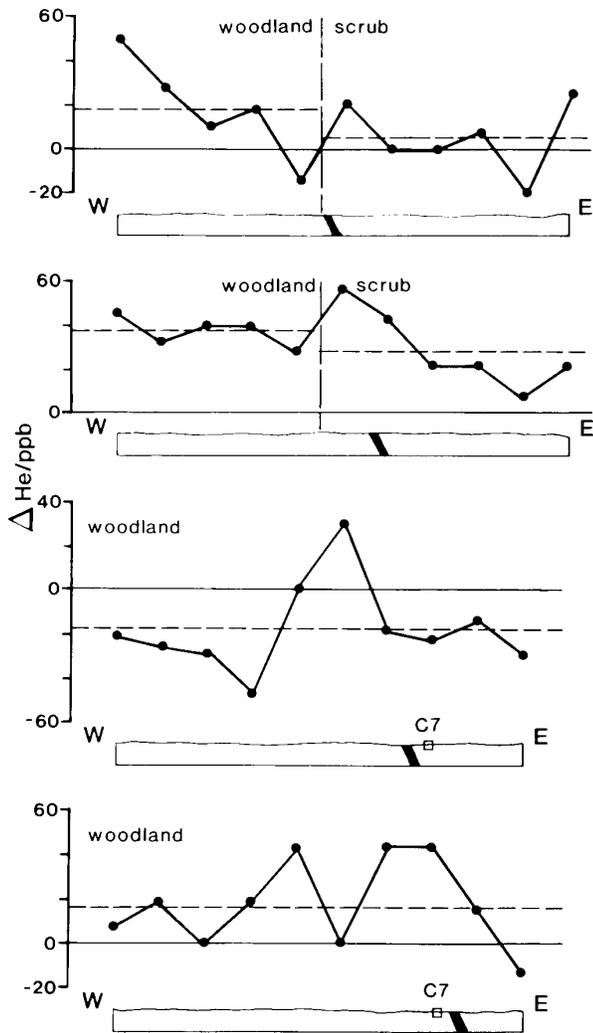


Figure 3. Soil gas helium variation on four E-W traverses across NNWSE trending U vein at Kingswood.

Durrance 1987a) indicate that ⁴He is as much a structural indicator as it is one of U mineralisation.

Discussion

Many authors have reported the use of dissolved trace gas concentrations and/or soil trace gas concentrations as specific pathfinders for mineralisation, fracturing, gas field or geothermal exploration. Some reports show strong positive correlations between trace gases (such as ⁴He and ²²²Rn), while others show only poor or even negative correlations (Gregory 1987). Similarly erratic behaviour may be seen for both dissolved gases and soil gases. The occurrence of such variable patterns in the relationship between gases in groundwaters is a strong indication that this behaviour is in part controlled by the geology, and in part by the groundwater hydrology. Consequently, the evolution of volatiles is obviously site-specific in character.

Gregory and Durrance (1987b) proposed a regional hydrothermal circulation model which accounted for varying rates of flow in fractures controlling dissolved gas release to the surface. However, the relationships seen above in the cases associated with U mineralisation led Durrance and Gregory (1987) to further consider the effect of gas solubilities as hydrostatic pressure decreased approaching the water-air interface, combined with the additional effects of high-solubility

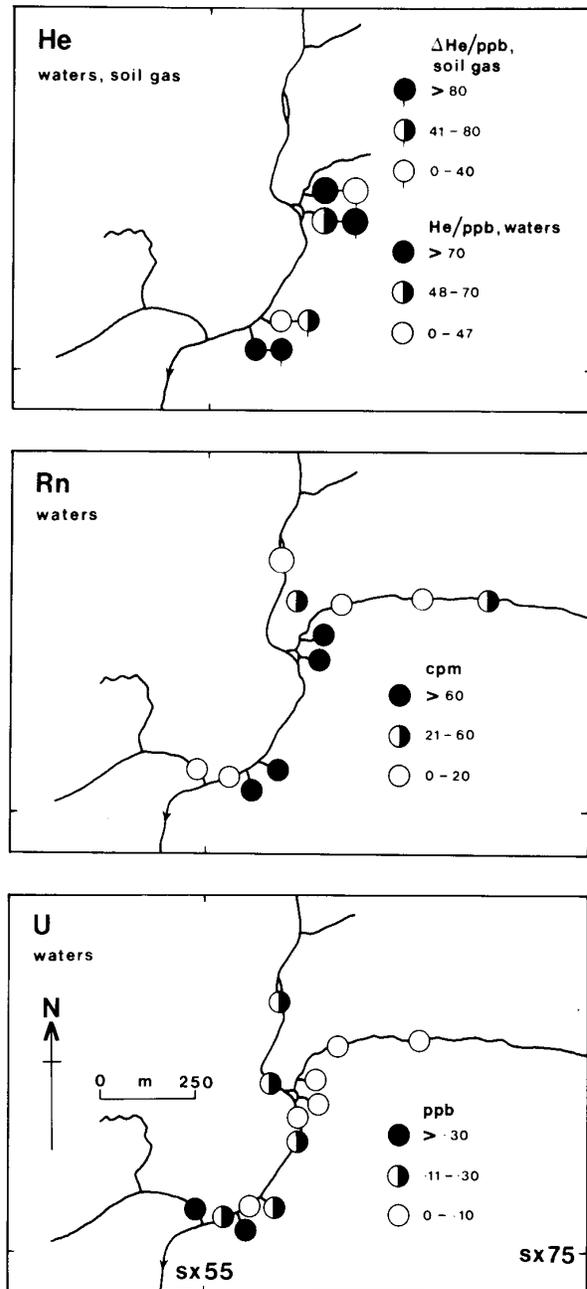


Figure 4. Soil gas and dissolved helium, dissolved radon and uranium at Merrivale.

oversaturated gases (such as CO₂) 'exsolving' and in part scrubbing low solubility trace gases with it. This model requires further detailed study, but supports the data from Merrivale, where soil gas ⁴He (rapidly exsolved) closely correlates with dissolved ²²²Rn. It is likely that both models are end-members of an infinitely variable groundwater - gas relationship.

It is a common observation that convincing soil gas ²²²Rn anomalies are usually seen over vein-type U mineralisation. Other soil gases, such as CO₂, O₂ and ⁴He, have also been used in the detection of mineralisation and fractures. Research carried out on ⁴He shows that this soil gas is not necessarily an indicator of U mineralisation. Rather it reflects the total flux of ⁴He from the degassing Earth. All soil gases are affected by near-surface effects, such as soil moisture and other pedological and meteorological variables, as well as deep-seated ones. This can lead to misleading patterns observed in soil gas studies. Such

variability in behaviour is repeated in studies of dissolved trace gases, and in the dissolved gas - exsolved gas relationship. In some cases, dissolved ^4He and ^{222}Rn directly reflect the concentration of U in solution. In others there is no spatial association with the chemical distribution of U or its daughter isotopes. Although there is evidence for a broad correlation between dissolved and exsolved gases on a regional basis, the coexistence of gases in solution and in the vapour phase is not a simple relationship.

Dissolved gases may be transported in solution only when the concentration is less than the maximum solubility under ambient conditions of pressure, temperature and salinity. In practice, excess gas concentrations may be transported wholly in solution, or as a combination of dissolved gases and discrete 'aerosol' phases. Those gases most likely to exsolve are those with low solubilities. Of these, ^4He has the lowest solubility.

In the upper crust, gas solubility combined with circulating groundwater flow systems control the degree of degassing from the liquid phase. In some instances, differential behaviour trace gases in solution and in the vapour phase occur. This may be related to the degree of mixing of dissolved gases from differing sources (deep-seated, radiogenic, or even recycled atmospheric). Consequently, large-scale (regional) gas anomaly patterns reflect the nature of the groundwater flow system. In contrast, smallscale (local) gas emanation is site-specific in character, and related to the local flux of terrestrial volatiles.

Conclusions

Soil gas ^{222}Rn measurements have the ability to clearly define radiometric anomalies, by way of their high anomaly/threshold values. Soil gas ^4He measurements do not. Where soil gas ^4He variations are observed, they are of comparable magnitude to those seen over other types of fracture, and merely represent the bulk flux of ^4He from the Earth. By comparison, the input of radiogenic ^4He from near-surface ores is minimal.

In waters, correlations of ^4He or ^{222}Rn with U are only justified on a local scale. Regional patterns of distribution reflect large-scale structural and hydrological controls. The solubility coefficients of dissolved gas species influence gas transport by advective processes as importantly as do absolute concentrations. Exsolution of dissolved gas by changes in temperature or lowering of hydrostatic pressure can result in scrubbing of lowsolubility gasses (such as ^4He) by gases with higher dissolved concentrations (such as CO_2).

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