Jointly published by
Elsevier Science S. A., Lausanne and
Akadémiai Kiadó, Budapest

Journal of Radioanalytical and Nuclear Chemistry, Articles,
Vol. 193, No. 2 (1995) 291–300

EVIDENCE FOR RADON TRANSPORT BY CARRIER GAS THROUGH FAULTED CLAYS IN ITALY

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Extensive soil-gas surveys in sedimentary basins in Italy were performed to study the potential of some naturally occurring gases as indicators for concealed fracture zones, hydrocarbon and geothermal fluids. One conclusive result is a positive correlation between anomalously high values of radon and carbon dioxide in the soil-air over faults. The correlation coefficient for 1173 gas samples is 0.41. Statistically derived contourlines of Rn and CO_2 anomalies show similar locations, shapes and directions. Fairly good Rn-CO_2 coupling evidence appears even on a point-to-point analysis. Furthermore, it was recognized that the highest Rn values are in contrast to the low Ra content of the underlying clayey rocks and that conventional Rn transportation mechanisms seem to be inadequate for the clay sequences. All these facts strongly suggest that Rn is transported from the subsoil, through fault-linked pathways, by carrier gases of which CO_2 could be one of the major components. The theory of geogas microbubbles is a possible explanation of the observed results. The carrier effect of ascending microbubbles can explain both the origin of soil-gas Rn anomaly and the Rn-CO_2 coupling phenomenon.

^{222}Rn (hereafter simply referred to as radon or Rn) measured on the Earth's surface is generated by ^{226}Ra decay in soil, water or outcropping rocks. Generally, radon produced in subsoil contributes to surface radioactivity if upward Rn transport by groundwater and gas diffusion occurs in the unsaturated zone[1]. Because of radon decay, such transport mechanisms, being quite slow, can explain Rn values at the surface only if U bearing rocks are located at very shallow depths. Nevertheless, during the last fifteen years evidence of long distance radon transport through geological formations has been recognised[2]. Consequently further migration mechanisms have been suggested. Mogro-Campero & Fleischer (1979)[2] studied radon transport by means of fluid convective movements due to geothermal gradient. Clements & Wilkening (1974)[3], proposed a radon migration linked to atmospheric pumping-driven flow. Later, Kristiansson & Malmqvist (1982)[4] suggested a new theory on long distance radon migration in which advective movement of carrier gas microbubbles through water-filled faults plays a key role. A theoretycal model for "microbubbles" transport was developed by Varhegyi et al. (1986)[5]. Durrance & Gregory (1990)[6] explained soil gas Rn anomalies using such theory. Their statement is that "rapid transport of ^{222}Rn from a greater depth than is possible in flowing groundwater is taking place by means of a carrier gas". They also considered CO_2, H_2, N_2, CH_2 and CO as potential carrier gases. Ball et al. (1991)[7] outlined close relationships between Rn and CO_2 anomalies in soil gas over rocks with low U contents and inferred that "rapid fluid transport along faults can give rise to anomalous concentrations of radon in soil gas remote from uranium and radium enrichments". They considered carbon dioxide as a suitable carrier for deep origin radon. Similar conclusions were reached by Etiope & Lombardi (in press)[8] studying radon and carbon dioxide distribution over clayey sequence several hundreds meters thick. In the present paper more extensive evidence of Rn transport by CO_2 through pelitic sequences in Italy are discussed. The Rn migration mechanisms, the role of carbon dioxide as carrier gas and geological features are considered in order to explain high Rn values in clayey sediments.

0236-5731/95/US \$ 9.50

Soil-gas surveys

Soil-gas studies based on the use of naturally occurring gases as geological pathfinders, have been extensively performed since the early '80s in Italy. More than 20,000 samples were collected all over Italy for hydrocarbon, mineral and geothermal exploration, fault mapping, seismic and volcanic hazard. One of the main findings was that soil air above active faults is sistematically enriched in endogenetic gases since, as known, faults are significant "leaks" for the earth's outgassing process.

In this paper only the results of surveys carried out in clay basins are discussed in order to point out a possible mechanism of Rn migration through very low U bearing (and scarcely permeable) formations. The case studies considered here refer to 6 sedimentary areas in central and southern Italy: Siena Basin; Medium Tiber Basin; Roveto Valley; Ofanto Basin and the petroliferous areas of Pisticci and Gela (Fig. 1). These areas are placed on tectonic deeps of various nature (graben, intramontane and foredeep) with pliocenic clayey covers whose thickness ranges from 100 up to 2000 m. The main litotype refers to homogeneous clayey rocks whose U content does not exceed 2-3 ppm (direct measurements of ^{226}Ra in the Medium Tiber basin exhibited a mean value of 2 ppm equivalent of U [9]). Even the sedimentary basement, underlying the clay covers, consists of rocks with very low U contents, such as limestones, marls and sandstones.

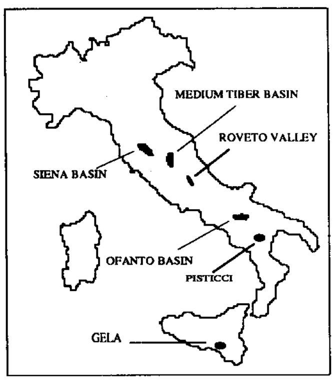

Fig. 1 Location of the studied areas

A total 1346 soil gas samples - of which 1173 include both Rn and CO_2 data - were collected and analysed. Soil-gas prospecting was performed on a regular grid network in areas ranging from 1 up to 200 km^2 and with a sampling density ranging from 1 to 400 samples per square kilometer, depending on the scale of each survey (regional and detailed). Each survey was carried out during dry season and in a short period to minimize variation of soil-gas concentration induced by climatic changes. Soil-gas sampling was conducted at depth of 0.5 meters in the soil according to a technique developed by the U.S.Geological Survey and described elsewhere[10]. Rn was analysed using a portable scintillation counter (EDA RDA 200) and CO_2 by means of a "quadrupole" mass spectrometer (VG SX 200).

Results

Table 1 lists some descriptive statistics of Rn and CO_2 concentrations in soil-gas samples. In the following discussion, for both gases, values above the mean plus 1/2 standard deviation were considered anomalous. In Fig. 2a, b, c, d, e, f, g, h, such anomalies are highlighted on

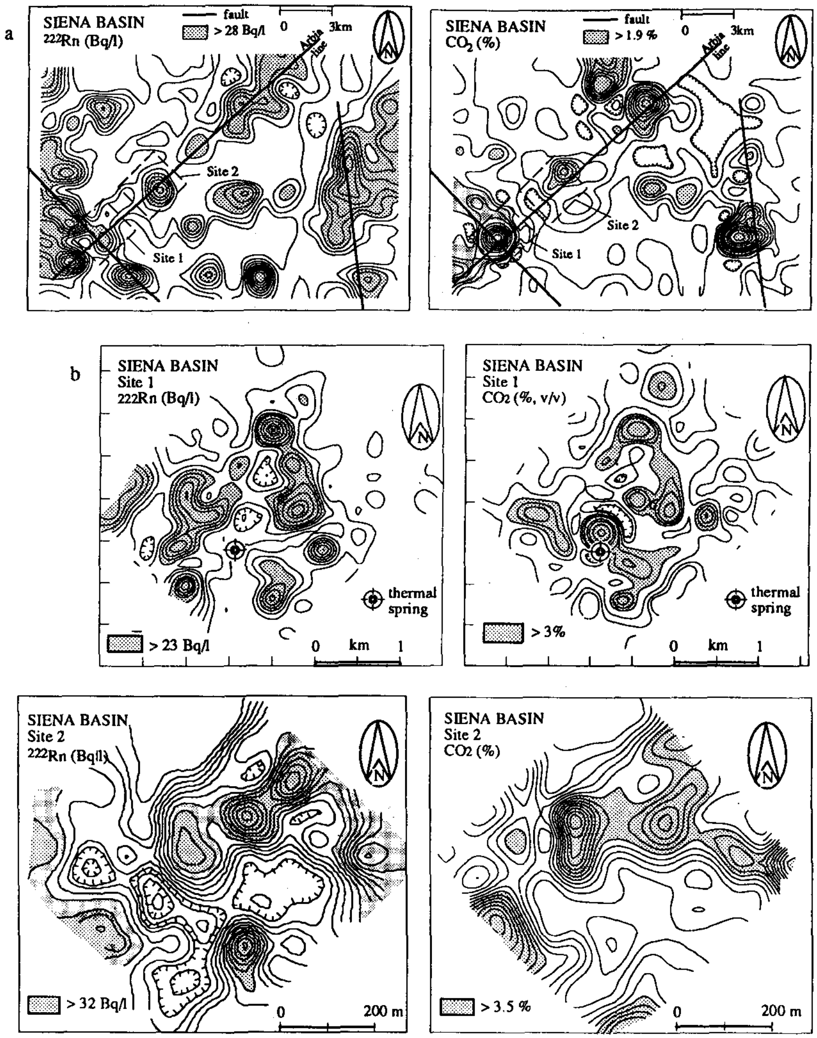

Fig. 2 a-c. Maps of Rn and CO2 soil-gas distribution in the Siena Basin. Contourlines are derived by "kriging" interpolation method. Anomalies are shown up by grey areas. Continuous lines represent faults reported in literature.

contourline maps of gas distribution obtained by computer-processed "kriging" method. On the same maps, fault lineaments reported in literature are shown [9, 11, 12, 13].

As depicted in Table 1, Rn activity in soils has mean values of the same order of the world-wide average (i.e. 7-18 Bq/l; [1, 14]), while the highest values reach levels of 70-110 Bq/l, unlikely supported by the mean local concentration of ^{226}Ra. The range of CO_2 values extends from ambient atmospheric values to levels three orders of magnitude above atmospheric, with the highest values above the normal microbial CO_2 [15]. Soil-gas anomalies are not randomly

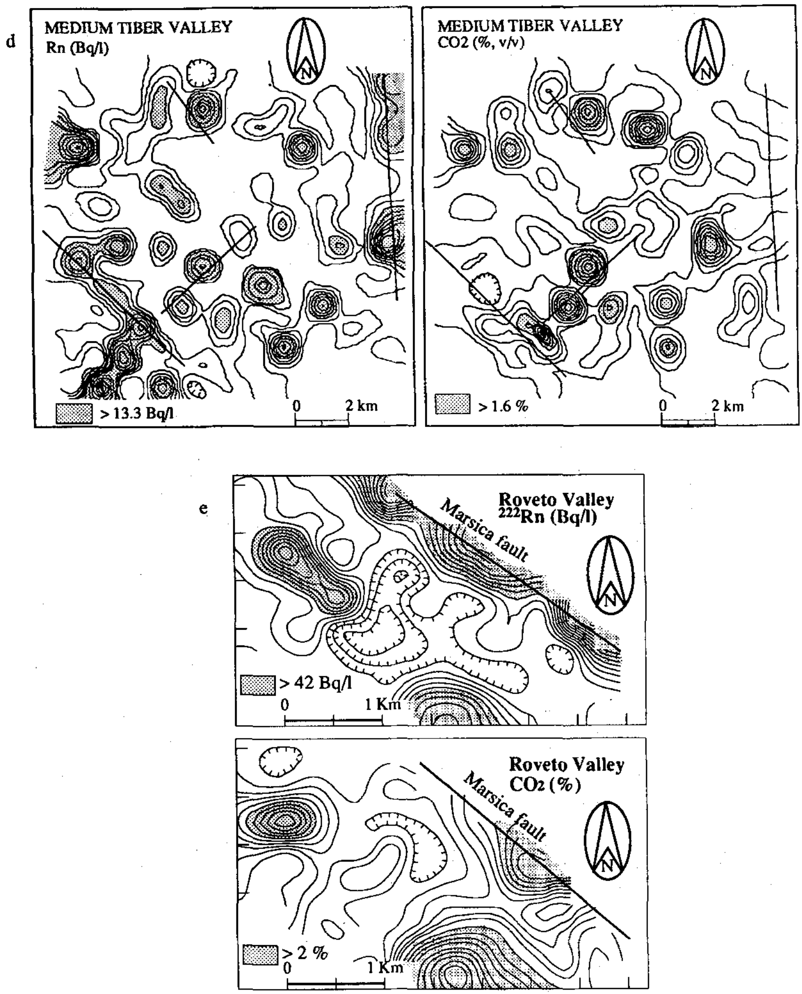

Fig. 2 d,e. Maps of Rn and CO_2 soil-gas distribution in the Medium Tiber Valley and Roveto Valley (Central italy)

distributed and preferentially occur close to or directly above faults and fractured zones. Most anomalies correspond to sites morphologically depressed and/or with evidence of photogeologic lineaments such as talwegs and scarps, i.e. morphological structures related to tectonics [8, 16].

A close direct correlation between Rn and CO_2 is evident in all the studied areas. Point-to point analysis showed that 64% of the CO_2 anomalies were detected jointly with a Rn anomaly. The CO_2-Rn coupling is evident also by comparing location and shape of gas anomalies on the contourline maps derived by "kriging" interpolation method. Surprising resemblance between Rn and CO_2 is exhibited by Site 1 and 2 maps of Siena Basin (Fig. 2 b,c). As known, even if soil gas measurements can be punctually affected by bias errors (induced by climatic factors,

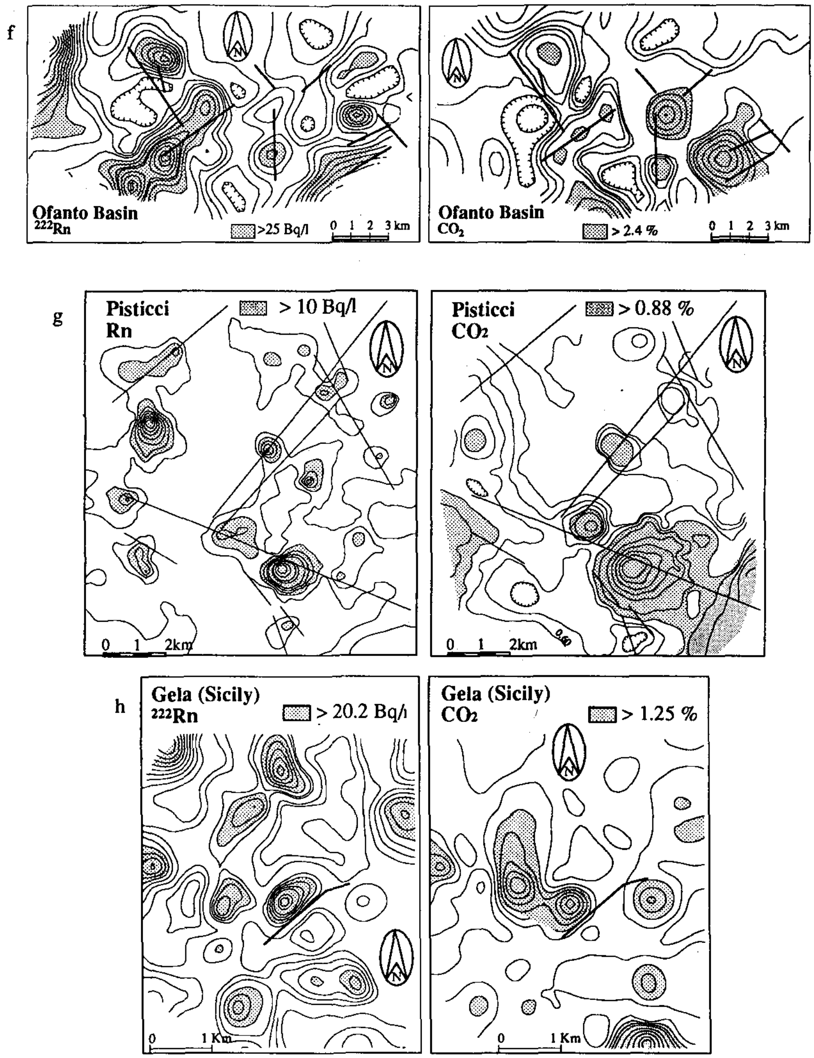

Fig. 2 f-h. Maps of Rn and CO_2 soil-gas distribution in the Ofanto Basin, Pisticci Basin and Gela area (Southern Italy)

instrumental errors, etc.) and erroneous interpretations can be given by biologic origin of CO_2, statistically derived contour lines of soil-gas concentrations should smooth such possible errors. Showing similar location, geometry and direction between Rn and CO_2 anomalies, the statistical approach effectively highlights the coupling phenomenon of these gases.

Table 2 shows the CO_2-Rn correlation coefficients. A coefficient of 0.41 has been calculated for the whole sample set.

Table 1
Descriptive statistics of Rn and CO_2 data

GAS	*Mean*	*Std.Dev.*	*Min.Val.*	*Max.Val*	*Count*
Siena Basin (regional sampling: 1 sample/ km^2)					
^{222}Rn (Bq/l)	20	15.93	0	90.28	249
CO_2 (%)	1.31	3.22	0.03	46	249
CO_2^* (%)	1.13	1.53	0.03	15.89	248
Siena Basin (Site 1) (detailed sampling: 25 samples/ km^2)					
^{222}Rn (Bq/l)	14.17	18.87	0	100.64	195
CO_2 (%)	2.77	4.72	0.30	58.25	195
CO_2^* (%)	2.19	1.85	0.30	8.55	188
Siena Basin (Site 2) (detailed sampling: 400 samples/ km^2)					
^{222}Rn (Bq/l)	23.88	16.63	0	71.78	81
CO_2 (%)	2.58	1.96	0.13	9.8	81
Medium Tiber Basin (regional sampling: 1 sample/ km^2)					
^{222}Rn (Bq/l)	8.83	9.14	0.74	61.05	258
CO_2 (%)	1.11	3.10	0.03	45.81	256
CO_2^* (%)	0.93	1.31	0.03	14.1	255
Roveto Valley (detailed sampling: 25 samples/ km^2)					
^{222}Rn (Bq/l)	32	21	3	112	100
CO_2 (%)	1	2	0.01	7	100
Ofanto Basin (regional sampling: 1 sample/ km^2)					
^{222}Rn (Bq/l)	16.76	17.26	0.74	97.31	110
CO_2 (%)	1.33	2.10	0.13	15.51	106
Pisticci (regional sampling: 2.5 samples/ km^2)					
^{222}Rn (Bq/l)	5.36	8.22	0	82.51	267
CO_2 (%)	0.50	0.76	0.03	4.80	66
Gela (regional sampling: 4 samples/ km^2)					
^{222}Rn (Bq/l)	12.68	15.27	0	77.33	86
CO_2 (%)	0.8	0.9	0.03	6.53	86

* Statistics computed discarding huge concentrations linked to thermal springs

Table 2
Correlation coefficients between Rn and CO_2

Siena Basin	0.40
Siena Basin (1)	0.56
Siena Basin (2)	0.68
Med.Tiber Basin	0.45
Roveto Valley	0.53
Ofanto Basin	0.21
Pisticci	0.87
Gela	0.58
Total	0.41

In the Siena Basin and Medium Tiber Valley some major gas concentrations were measured around visible gas emissions from the ground; at some sites water saturated ground (close to springs, irrigation channels or river beds) gas bubbles were seen bursts.

Finally, we observe that during some of the soil-gas surveys the helium content was analysed [8, 9, 12, 16] and that concentrations well above the atmospheric level (i.e. 5220 ppb) were detected in the same fracture zones characterised by both Rn and CO_2 anomalies. The occurrence of helium anomalies implies that those fractures act as pathway for deep origin gases[17]. Non-coincidence on point to point basis among anomalies of different gases is common. Hence, the observed Rn-CO_2 coupling is somewhat surprising and, considering the large number of data, it assumes an important meaning for the interpretation of the gas anomalies themselves.

Discussion

The above results show a remarkable Rn-CO_2 coupling and the occurrence of high Rn values for clay with very low Ra content is unanticipated. The interpretation of such results requires a review of the literature.

To explain the observed fault-linked radon anomalies, the occurrence of high emanation coefficient in faulted rocks must be taken in account, as it usually provides a significant contribution to the soil-gas Rn level. Nevertheless in the studied areas calculation (using emanation coefficient, porosity, density and Ra content of soil) shows that enhanced emanation coefficient of clay cannot alone account for Rn values above 70 Bq/l, because of the low ^{226}Ra content of the clays. Hence, although localised Ra enrichments may exist (e.g. due to co-precipitation on soil grains) a further contribution from depth is generally required to support the highest radon values. To explain Rn contribution from subsoil several gas migration mechanisms in faulted rocks are considered afterwards.

One possible Rn source in subsoil, very well described for crystalline rocks[18], may be related to Rn transport by greater volumes of groundwater flowing through faults or to enhanced level of Ra concentration in the water itself. It is unlikely that this mechanism may apply to the studied cases since clays, even if fractured, usually have very weak water conductivity. If water movement occurs it is generally extremely slow and Rn transport by groundwater flow can be totally negligible. However, water-linked Ra enrichments along fractures cannot be disregarded, although they should be of slight importance. Anyway, frequent surface gas manifestations exist over faults in the studied areas [9,16]; therefore a more important cause of the high values of Rn over faulted clays, in most cases, could be linked with a gas phase migration along the faults themselves.

It is well known that diffusion alone cannot account for surface Rn anomalies where distance between Rn source and collection is much larger than a few meters. For longer distance and in more permeable zones, such as faults, advective upflow induced by pressure driving forces is the most important mechanism of volatile migration. Two interesting hypotheses on gas upflow regard convection by geothermal gradient [2] and atmospheric pumping-driven flow [3]. They do not seem to be adequate for the clayey basins, however, since these processes usually provide insufficient driving force[2,3] and require special favorable conditions, such as very permeable soil and overburden and extraordinary thermal gradients, which do not exist in the studied areas. To explain the observed Rn anomalies a different advective Rn upflow is required. It must be noted that to move by advection, i.e. to be sensible to gravitative forces such as pressure gradients, a gas must have a sufficient amount of mass (it must form a "gas domaine" as suggested by Gold & Soter, 1985 [19]). To form a stream of pure radon an immense number of Rn atoms must be available at the same spot at the same time. The amount of radon in the underground environment (orders of 10^{-10} ppm) is too low to form a macroscopic quantity of gas which can flow advectively through the geological formations. Therefore Rn must be carried by a flow of another gas which is moving upward. Some authors [6, 7] suggested that carbon dioxide, being quite abundant in the subsurface environment, is one of the most suitable carrier gases for Rn. In the areas of Gela and Pisticci CO_2 flow is probably linked to the occurring hydrocarbon reservoirs whereas in the Siena Basin it can have relationships with low enthalpy geothermal fluids existing below the clayey cover [11]. In the other areas, to directly recognise the possible sources of carrier gas (e.g. CO_2) is quite difficult. Nevertheless, in Ofanto basin, in Medium Tiber Valley and in Roveto Valley oil fields, thermal fluids and a very deep fault system, respectively, may be inferred. However many authors [19,20,21,22] suggested that a micro-flux of gas may also exist regardless of the occurrence of localised source. So, transport of Rn by naturally migrating carrier gas seems to be an important way in which soil-gas anomalies originated.

As microbubble streams can form during escaping of terrestrial gas through groundwater-filled fractures [4,5,23,26], the role of geogas microbubbles on Rn transport, as suggested by several authors [4,5,6,21], cannot be disregarded. Evidence of rising bubbles in different geological environments are recorded throughout the world [4, 22]. The formation of bubbles in the subsurface requires gas oversaturation or may be aided by some kinds of mechanisms that reduce the activation energy of formation (for example ionizing radiation emitted from radioactive elements in the fracture coatings [23]). Oversaturation may be induced by local pressure shocks or by local overproduction of gas (for example the CO_2 produced from the oxidation of anoxic C where deep groundwaters meet shallow oxic groundwaters [6]). The movement of the bubbles is due to the difference in density between the bubble and the water in accordance with the Stokes' law [5]. The bubbles ascend along faults until they reach the groundwater surface; the bubble gas is then mixed with the soil-overburden gas. The gas mixture so formed could be slowly forced upward by the pressure gradient caused by the bubble stream [4].

The movement of microbubbles through geological formations may give rise to enhancement of transport of dissolved gases from the groundwater (water phase) to the bubble (gas phase) and subsequent their transport upward. Microbubbles could also pick up matter (gas atoms as well as solid microparticles) from rocks and to lift it upward by particulate flotation[24], transport of aerosol inside the bubble and binding of surface active elements onto the gas-water interface [25]. Through such mechanisms, lifted matter can "heap" on the bubbles (or inside them) and be released and concentrate where bubbles burst. It was experimentally proved that radionuclides can potentially be involved in such processes. Enrichments of Cesium [25], Radon [26] and Plutonium [27] were observed on water surfaces after bubbling water with air. There are no experimental data known to the authors on Ra and U transport by bubbles. However, it is plausible to suggest for them the potentiality of this kind of migration. Even if there is not a direct evidence of radionuclide transport by rising microbubbles, such transport phenomena could effectively explain the occurrence of Rn anomaly over low U bearing rocks, such as the pliocenic clays, and the Rn-CO_2 coupling phenomenon. Because of the mass impermeability of clays, no doubt microbubbles streams must take place exclusively through fracture networks.

So, it is possible to suggest that geogas flow does not transport Rn from localised or very deep sources (e.g. in the mesozoic bedrock which can be up to two thousands meters depth), but that during migration upward through faults microbubbles pick up Rn atoms emitted by the normal content of Ra of clayey rocks and/or pick up directly its parent radionuclides (Fig. 3).

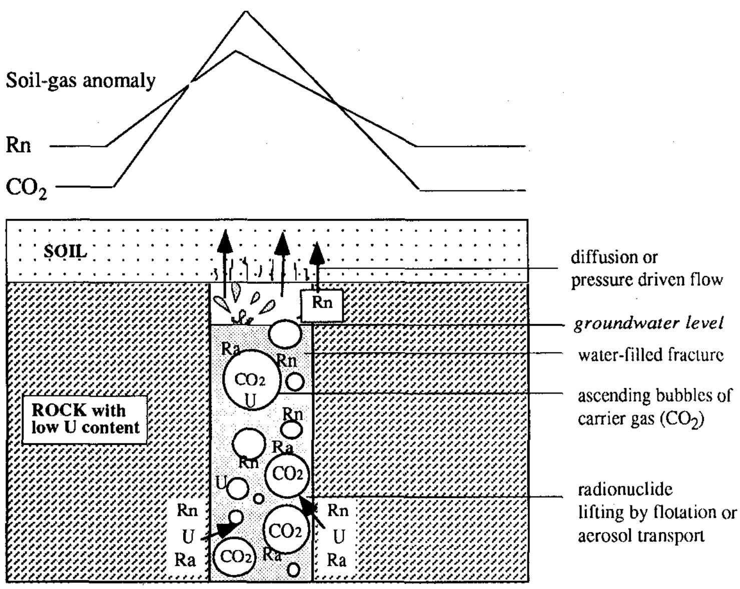

Fig. 3 Sketch model of generation of Rn-CO_2 coupled anomaly induced by Rn (and/or U and Ra) transport by carrier gas microbubbles through a fault in low U bearing rocks.

Little by little geogas microbubbles get enriched with radionuclides drawing them from the several geological horizons. When microbubbles reach the groundwater surface they burst and thus radionuclides are released. Hence, radon migration by diffusion and/or advection (depending on the gas total pressure developed in the pores above the capillary firinge) towards the soil occurs. In the case of Ra or U lifting, as such elements have a long half-life, no particularly high migration rates are requested to product soil-gas Rn anomalies. If carbon dioxide is one of the components of gas bubbles, such a "heap transport" process would explain the strong correlation between Rn and CO_2 at surface.

Basically, it is worth noting that such a process requires neither the occurrence of any specific or localised Rn source nor huge migration velocities essential for admitting a Rn origin from very deep U bearing rocks or fluids. Obviously, this hypothesis needs experimental validation.

Concluding remarks

The results from extensive soil-gas surveys in clay basins evidenced the occurrence of high Rn values unexpected for clay with very low Ra content and a remarkable Rn-CO_2 anomaly coupling over fault zones. A Rn contribution from the subsoil was thought necessary, but conventional Rn migration mechanisms did not seem to be completely adequate for the existing boundary conditions. On the contrary the geogas microbubble theory well lends itself to explain all the observed results. The carrier effect of ascending microbubbles allows an explanation for both the origin of soil gas Rn anomaly, due to radionuclide transport by rising bubbles, and the Rn-CO_2 coupling phenomenon, inasmuch as carbon dioxide is the most suitable carrier gas in the considered geologic environments. However, the importance of microbubble flow in the geological environment has yet to be assessed.

This process might not necessarily correspond to the most important gas migration mechanism in the subsurface. Furthermore, some surface Rn anomalies may be linked to localised Ra enrichments. Nevertheless the potential for radionuclide transport by microbubbles exists and, basically, it provides a real new research perspective on radionuclide behavior and migration in the geosphere.

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The authors would like to thank Mr. E. Grillanda and Mrs. A. Baccani for technical support during soil-gas surveys. Part of this research was supported by EC funds (Contracts N° F12W-0064 and N° F12W-0121).

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