

AN INTERCOMPARISON OF SEVERAL METHODS OF DETECTING RADON-222 IN OVERBURDEN GASES¹

by

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(8 figures and 2 tables)

RESUME.— Les résultats obtenus en utilisant quatre différentes techniques de détection du Rn222 dans les gaz du sous-sol ont été comparées. Trois techniques intégrées et une technique instantanée (émanométrie) ont été employées. Les techniques intégrées étaient basées sur l'utilisation de la thermoluminescence naturelle des minéraux des formations superficielles, de la thermoluminescence de matériaux artificiels placés sur le site (TLD), de l'absorption passive de radon sur du charbon actif et de deux types de films sensibles aux particules alphas. Le site test est situé à Blaton (Belgique) et montre une couverture de sables tertiaires reposant sur des formations paléozoïques contenant quelques faibles concentrations en uranium. Une bonne correspondance a été obtenue entre toutes ces techniques dans la localisation des anomalies en radon dans la couverture.

ABSTRACT.— An intercomparison under field conditions was made between four different techniques of detecting Radon-222 in overburden gases. Three integrating and one instantaneous technique (emanometry) were used. The integrating techniques were based on the use of : - natural thermoluminescence of minerals in the overburden, artificial TL material placed there (TLD), passive adsorption of radon by activated charcoal, and two types of etchable alpha track plastics. The test site used is in Blaton (Belgium) where tertiary sands overlie paleozoic formations with some weak occurrences. Good agreement was obtained between all the techniques in locating the principal radon peaks in the overburden.

INTRODUCTION

The study of the radon distribution in the soil leads to various applications : prospecting for buried deposits of uranium and phosphates (Smith, Baretto, Pournis, 1976), earthquake prediction (King Chi-Yu, 1978), exploration or monitoring of geothermal systems (Kruger, Stoker, Umana, 1977 ; Whitehead, 1981) and prospecting for petroleum (Armstrong, Wood, 1973). With the support of the EEC Community (1) (and of the "Programmation à la Politique Scientifique", Belgium) research has been carried out by University College Dublin (Ireland) and the Faculté Polytechnique de Mons (Belgium) in order to compare several techniques of radon detection in superficial formations. Six different methods were applied under the same field conditions : natural thermoluminescence of the minerals of the superficial formations, TLD (thermoluminescence dosimetry) detectors, activated charcoal detectors, alpha track plastic detectors, emanometry and absolute radon measurement by sampling gas in the soil. This intercomparison was made in the Blaton

area in Belgium near Mons. This site afforded the possibility of studying in good sampling conditions the radon distribution in an area with some rather weak uranium anomalies.

DESCRIPTION OF SITE

The Blaton site had been used by the Geophysical Group of the Faculté Polytechnique de Mons to test various exploration techniques (ground and airborne radiometric methods, electrical methods, . . .). It also

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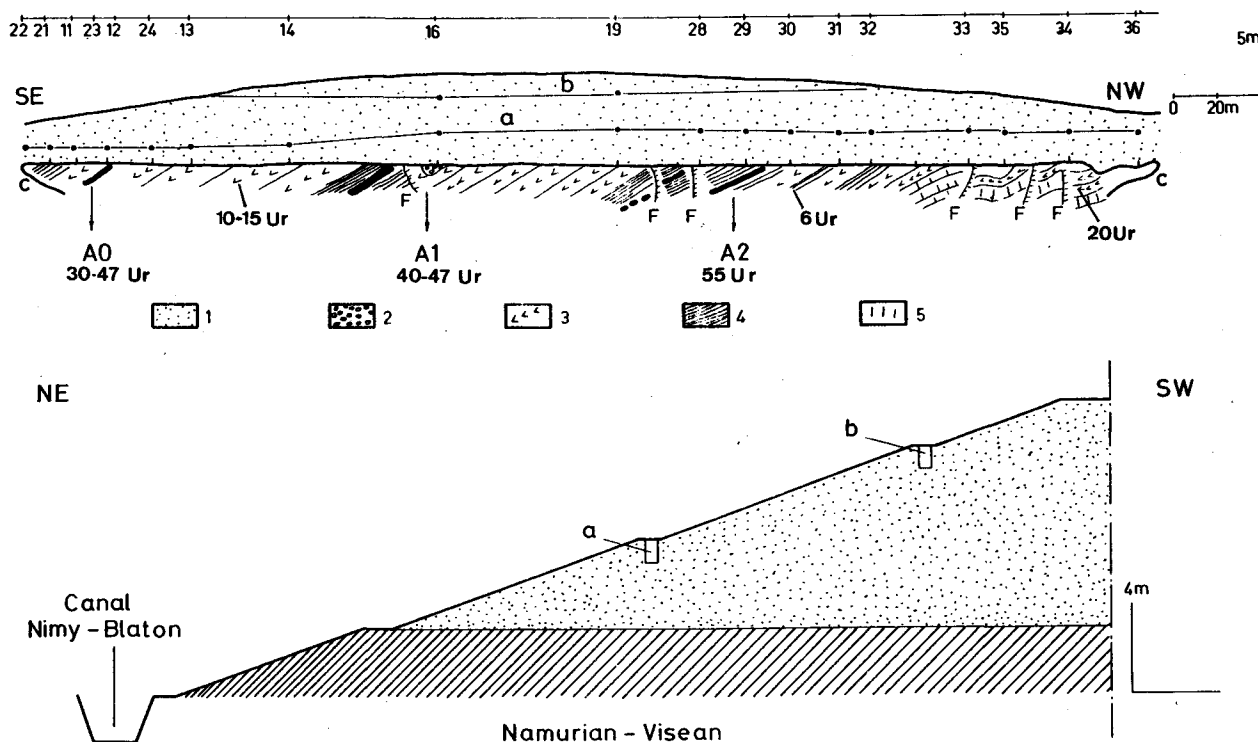


Figure 1.- The Blaton test site, general features :

a) lower path ; b) upper path ; c) path along the underlying formations.

1. overlying sands - 2. cretaceous conglomerate - 3. silicite - 4. black shales - 5. limestones.

appears to be an interesting zone to test radon gas detection methods in the overburden for several reasons. As can be seen in Figure 1 tertiary sands (Landenian) overlie in unconformity older geological formations with some uranium occurrences. They are both very well exposed along the Nimy-Blaton canal, an important navigable way in Belgium.

Along the towing path of the canal (Fig. 1) several uranium occurrences have been detected by a gamma-ray survey and by a new radon technique developed in Mons laboratory (Charlet, Dupuis & Quinif, 1977 and 1978).

These occurrences are as follows :

- a Cenomanian conglomerate (anomaly A1) of paleo-channel shape quite below the unconformity surface between the Paleozoic formations and the Tertiary sands,
- some radioactive beds (anomaly AO) distributed in a few meters of thickness in a silicite formation for which the precise age is not well known (Namurian or Visean).

- a radioactive zone (anomaly A2, of thickness : a few meters) located in the Upper Visean shales formations. It reoccurs to the southern part of the geological section at several places as a result of a fault system but to the northern part it finishes as a lens shape.

An extensive study of the anomalies of the host rocks has been carried out by radiometric survey and gamma spectrometry in the laboratory. The thorium and potassium percentages are very low ($Th/U < 1$, $U/K_2O > 10$ for the radioactive zones). The uranium grades (in radioactive equilibrium) are about 40 → 70 ppm. Therefore, the gross count data converted to the radioelement concentration unit (2) are in the field a good measure of the eU grade (fig. 1).

Overlying the paleozoic rocks, the Landenian sand formations form the hill named "Mont des Gro-

(2) A Ur is the count rate obtained for an infinite source containing one part per million uranium in radioactive equilibrium.

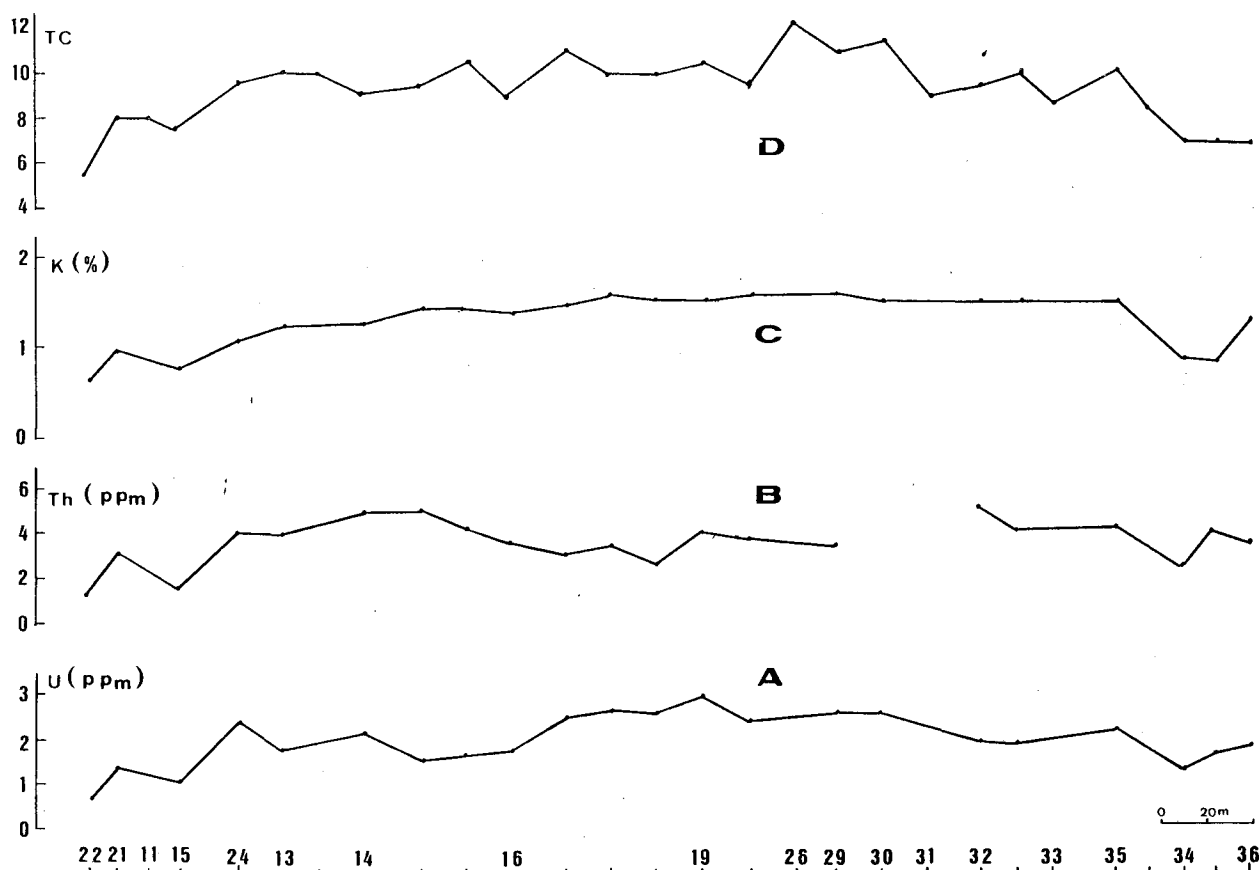


Figure 2

Radioelement content of the overburden sands, Blaton test site. A-B-C : results of the laboratory analysis by γ spectrometry (uranium equivalent in ppm, thorium in ppm, potassium K_2O in per cent). D : total count determined in the field with a scintillation detector N115B, measurements performed inside holes drilled with a screw-auger (in μ roentgen/h).

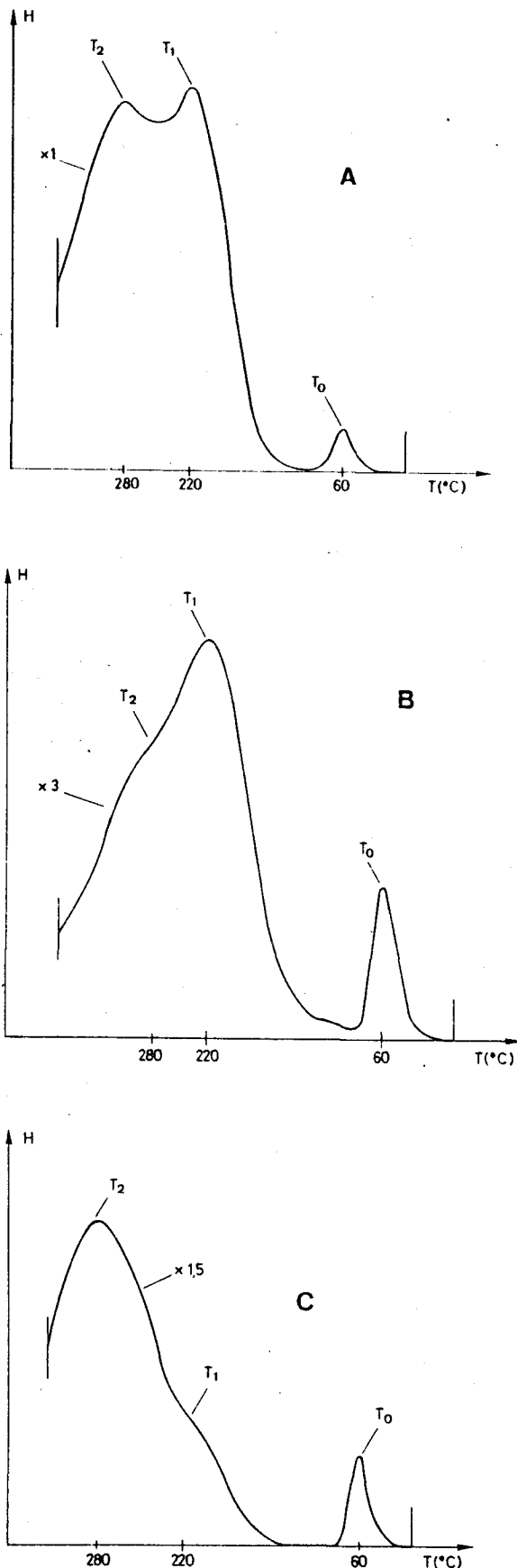
seillers" with some horizontal foot-paths along the side of the hill from which one can perform sampling and field measurements of the overburden in good conditions. A mineralogical study of the sands gave their feldspathic characteristics (a few per cent of plagioclases and K-feldspars). These features have an important role in the interpretation of T1 data.

A granulometric study of the sands shows an evolution from argillaceous sands at the bottom (grain size 80μ) to medium - or coarse sands at the top (grain size : 150μ). On the North and South side of the hill some sands have a bimodal granulometric distribution. They overlie the normal sands and are probably related to a flowing of the sediments (Charlet, Dupuis & Quinif, 1979) from the top to the bottom of the hill, giving rise to a mixture of fine and coarse sand. Along the profile where we have performed our sampling and our field measurements the thickness of the sands

varies from three meters to four meters. They are rather argillaceous with sands of a bimodal distribution on the right and left of the profile. Figure 2 gives the radioelements distribution for the profile examined. It can be seen that there is a good correlation between the grain size parameter and the radioelement content of the overburden sands. Where the sands are less argillaceous (at both ends of the profile), the radioelement content decreases.

In short the geological and radiometric study of the Blaton section allows us to distinguish several zones quite different with regard to the possibilities of radon migration of accumulation.

For the underlying formations two zones in the central and South part of the geological section appear as possible sources of radon represented by the uraniumiferous anomalies A0 and A2, the latter occurring at a



fault system. One can also include the anomaly A1 in relation to a cretaceous paleochannel.

For the overlying formations where the sampling was carried out on the horizontal path along the site of the hill the strongest variations for the uranium grade and thus for the radon coming from the sands appears at the ends of the profile in the rehandling zones. An extensive central part with a constant uranium content can be thus distinguished from northern and southern parts with a lower radioelement content.

DETECTION TECHNIQUES

A) NATURAL Tl OF THE SUPERFICIAL FORMATIONS

In 1977 the F.P.Ms. laboratory of Mineralogy proposed a new radon method for uranium exploration (Charlet, Dupuis & Quinif, 1977). It is based on the utilisation of a natural detector, the minerals of the superficial formations and thus on the recovering of a dosimeter that nature has deposited in the site a few millions years ago. The radiation effect in relation to the migration of radon may be revealed by thermoluminescence (Tl). A few years ago (Charlet, Dupuis & Quinif, 1978) a preliminary Tl study located several new anomalies at the Blaton site (anomalies A1 and A2) by the use of this technique. Complementary data is presented in this paper. We have investigated the Tl properties of the detrital minerals of the Landenian sand which can be considered as natural detectors (quartz and feldspars). Because the feldspar Tl intensity is stronger than the quartz Tl intensity (Fig. 3) it is often necessary to separate from the whole rock the interesting minerals phases. As the Blaton sands are not very feldspathic the extraction process of the K-feldspars is very long and a large sample of material (about two kilos) has to be used. It has often been necessary to restrict the Tl study to an assay from the whole rock (quartz and feldspars) and from the quartz. However the studies from the whole rock are only possible if the feldspar percentage remains constant at the local or regional scale. This was first checked in an earlier

Figure 3.

Natural thermoluminescence of the Blaton sands.

A : glow curve of the whole rock

B : glow curve of the K feldspar

C : glow curve of quartz

H : relative intensity (in arbitrary unit).

study (Charlet, Dupuis & Quinif, 1978). The experimental factors used in this study are given below :

- granulometry : 80-100 μ determined from the maxima of the frequency histogram,
- weight sample : 0,4 g of an internal temperature standard,
- heating rate : 1°C/sec (the duration of an assay is about ten minutes).

The TLD apparatus used has been already described in several papers (Charlet, 1969 ; Baleine, Charlet & Dupuis, 1973). The parameters used for the interpretation of the experimental data are the glow peak intensities H_{1n} and H_{2n} (3) (for the glow peak temperatures T1 and T2) and a glow curve shape parameter R defined by the ratio

$$\frac{H_1}{H_1 + H_2} \times 100$$

B) TLD (THERMOLUMINESCENCE) DOSIMETRY WITH CALCIUM FLUORIDE DETECTORS

In order to discuss the TI results of the overburden formations some different TLD detectors were used. The advantage of this system in the study of the radon migration zone during a period of a few weeks or months is that the same TI reader may be used as the one used in natural TI studies. Various TI dosimeters systems have been prepared by the Mons laboratory. For the field work on the Blaton site a system was used with a CaF_2 powder supplied by the company (Brussels, Belgium). After removing its natural TI by a heating treatment the CaF_2 powder is suspended inside small plastic cups. The glow curve of the natural calcium fluoride after irradiation with an artificial source exhibits several peaks. The low temperature glow peaks are strongly dependent on the "fading effect" in relation to the thermal fluctuations in contrast to the high temperature glow peaks which are strongly dependent on the effect of irradiation. With a view to using the two high temperature glow peaks and to smooth the data we calculate a parameter in which both the peak intensities are given the same weight. We take

$$H = \frac{(H_3 + 10H_4)}{4}$$

because the intensity H3 is approximately equal to ten times the intensity H4.

C) EMANOMETRY

Assays were carried out with a portable radon-content-meter (Saphymo-Stel Type EPP10) which utilises a $\text{ZnS}(\text{Ag})$ scintillation detector operating at constant flow. For a gas having a given radon concentration, the counting rate obtained by this method becomes nearly constant after a few minutes of pumping. The background count rate increases gradually due to memory effects from previous samples. Unfortunately it is not possible to convert the count rate into absolute radon units (pCi/l). As the radon activity is dependent on the climatological conditions a survey with this instrument was made several times before and after the intercomparison field period in order to obtain a measure of the mean radon content of the soil gas at the sampling locations.

D) CHARCOAL DETECTORS

Activated charcoal has been used for many years for adsorbing radon from air drawn through it, usually at low temperatures. It can also be used to passively adsorb radon even at ambient temperatures (Countess, 1976). This approach forms the basis of the field technique used in the present work. Small cylindrical plastic probes containing 20 g of special activated charcoal are buried in the ground at a depth of 0.5 to 1.0 metre for a typical period of 1 to 4 weeks. On recovery they are sealed and the gamma activity arising from the decay of ingrown radon daughter products (usually Bi-214) in the charcoal is measured using a NaI (TI) scintillation system. This gamma activity corrected for decay and background is proportional to the adsorbed radon. This inexpensive and easy to use technique gives an integrated measure of the radon at probe locations in the overburden. The particular version of this method used in the Blaton survey was developed in University College Dublin under an E.E.C. Contract. A similar technique is also commercially available. An ACRONYM CARP (charcoal adsorbed radon prospecting) is used to describe this technique.

E) ALPHA TRACK PLASTIC DETECTORS

The cylindrical probes containing activated charcoal also contained two types of alpha track plastic detectors. These were mounted facing the open end of the probes and therefore they were in a position to be

(3) Normalized by means of a standard to take into account the drift of the apparatus.

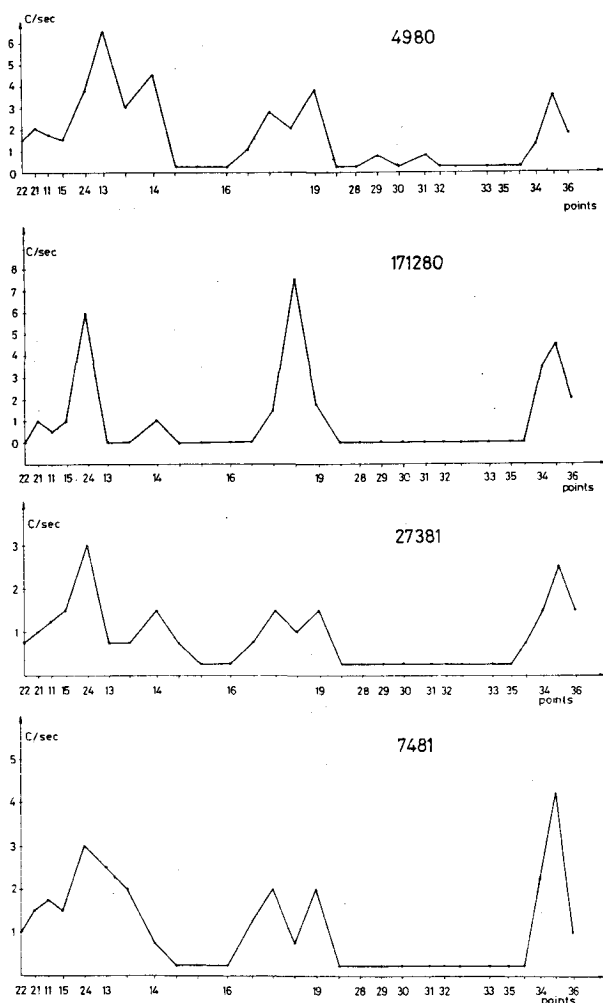


Figure 4

Some "radon" emanometry profiles carried out in 1980-1981 along the lower path of the hill "Mont des Groseilliers".

exposed to the alpha particles from radon and its daughters Po-218 and Po-214 when the probes were buried in the ground. The plastics when recovered were chemically etched in the Dublin laboratory. The alpha tracks so obtained were then counted using optical microscopy. The two plastic types used in this investigation were Kodak Pathé Cellulose Nitrate LR115 and a "new" type of very sensitive plastic called CR39 (Cartwright, Shirk & Price, 1978). For the Blaton series of exposures as may be seen in Figure 5, the CR 39 gave approximately twice the track density obtained using LR115. With the plastics in position the charcoal probes, therefore gave three integrated

signals due to radon exposure, a) the gamma activity from the charcoal, b) the LR115 alpha track density and c) the CR39 alpha track density.

INTERCOMPARISON

A) PROCEDURE

During the summer of 1980 field measurements were made at the Blaton site along the profile previously studied using TI (Charlet, Dupuis & Quinif, 1978 and 1979). Twentyeight holes of about 40-50 centimeters depth were dug in the overburden sand. Each of these holes was separated from its neighbours by a distance of about 20 meters. The original TI sampling points and also intermediate points were used with a view to obtained a more regular grid line.

On 22 July 1980, the following detectors were placed simultaneously at the bottom of the holes : TLD calcium fluoride detectors (Mons), charcoal detectors (Dublin), alpha track plastic detectors of types LR115 and CR39 (Dublin). Afterwards the holes were backfilled with sand. In addition some gas samples were taken from the soil for laboratory analysis in Dublin to measure absolute radon concentrations. The detectors were buried for a period of 43 days in the ground. They were recovered on the 4th September 1980 and the charcoal as well as the alpha track detectors were sent by air to Dublin the next day. In only one location a probe had been stolen and consequently twenty-seven sets of detectors were studied at the Dublin and Mons laboratories.

Since this joint field work at Mons, new studies have been carried out by natural thermoluminescence. Concurrently with the recovery of the detectors in September 1980 we took samples of the sands at the bottom of each hold at about a depth of fifty centimeters for the next series of natural TI assays. In addition in May 1981 a sample of sand was taken using a motorised screw auger at depths of about ten centimeters, two meters and three meters. In this way it will be possible to distinguish the effect of superficial disturbance and the relationship between the sampling depth and the natural TI level.

B) RESULTS, DISCUSSION AND INTERPRETATION

Emanometry field measurements were taken over a period of one year (from May 1980 to May 1981) to take into account the effect of climatological conditions on radon concentrations. All the profiles (Fig. 4)

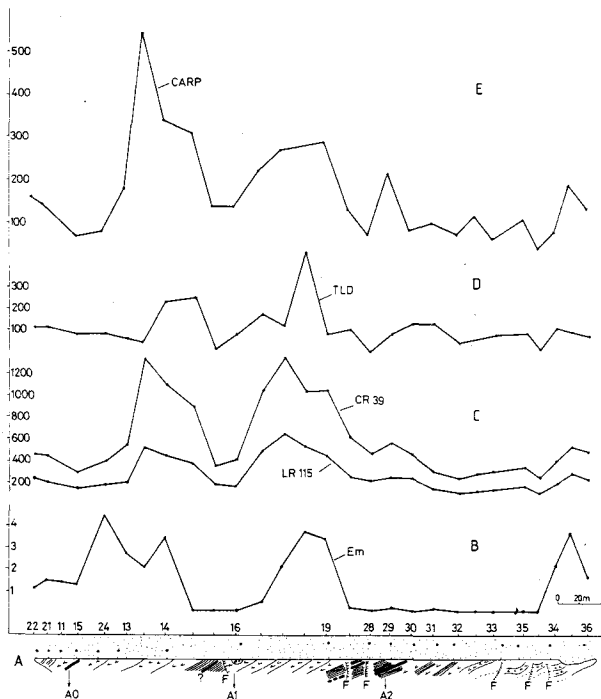


Figure 5

Intercomparison of Radon Detection Techniques :

- emanometry (Em),
- alpha track plastic detectors (LR115 and CR39),
- TLD (thermoluminescence) detectors,
- activated charcoal (CARP)

A : Geological section

A0-A1-A2 : uraniferous anomalies in Visean-Namurian formations or cretaceous paleochannel covered by "Tertiary Sands".

B : Emanometry (count/sec)

Average of the results from July to December 1980 to make up for climatological variations

C : Alpha track plastic detectors (tracks/mm²),

D : Thermoluminescence dosimetry (TLD) with calcium fluoride detectors (arbitrary unit)

$$\text{parameter } H = \frac{H_3 + 10H_4}{4} \quad \text{corrected for background.}$$

E : Activated Charcoal detector system (total gamma count rates corrected for background).

show the same shape :

- a peak between the points 34 and 36 having a very variable intensity,
- a zone of about 150 to 180 meters (from point 34 to point 28) with a very weak radon concentration,
- between points 28 and 22 a complex zone exists with several distinct radon maxima.

These maxima sometimes exhibit a slight shifting of position probably due to changing climatological conditions. It was, always possible to distinguish one or both of the peaks between the points 19 - 16, the minimum between the points 14 - 16, and one or several of the maxima between points 14 and 24. It is therefore possible to distinguish between :

- a northern area with a weak radon concentration except for the peak at the end of the profile (between the points 34 - 36),
- a central and southern area characterised by several zones of radon maxima.

This distribution seems to correspond quite well with the features established from an examination of the geological and radiometric data. The radon maxima are however, shifted, in comparison with the location of the anomalies A1-A2 (Fig. 5). It can be seen in Figure 5 that there is a general good agreement between the integrating radon techniques and the emanometry approach. It is necessary, however, to note that the integration times and the anomaly to background ratio were not the same for the different radon techniques (Pacer & Czarnecki, 1980). Thus, it is not surprising that there are some variations between the profiles shown in Figure 5.

The values of the absolute radon level obtained for the gas samples taken in July 1980 are given in the following table (Tab. 1).

| Points | 14 | 19 | 32 | 34 - 36 |
|-------------------------------------|------|------|----|---------|
| Concentration (in picocuries/liter) | 1050 | 1039 | 63 | 166 |

Points 14 and 19 correspond to the two main maxima of the emanometry profiles (Figs. 4-5), point 32 to the base level and the point 34-36 to a weaker and variable radon peak.

The values given in picocuries per liter appear rather low. From emanation theory (Alekseev, *et al.*, 1959) the radon level for the landenian sands at a sampling depth of 50 cm would be about 200-300 pCi/l. This is based on the radium content, (determined by gamma-ray spectrometry at the F.P.Ms. laboratory) the density and the porosity of the sands and on typical values of the emanation coefficient for sands. The application of emanation theory, therefore, suggests that the values of some hundred pCi/l are in relation to the radioelement content of the overburden sand and not to the underlying strata. However, if we consider that the radiometric uranium (eU) content of the superficial formations can be regarded as a good measurement of the local source of radon the bad correlation (compare Figs. 2 and 4) between eU concentration of the sands and the results of the radon surveys suggest the existence of some stronger sources of radon than the sands. The concentrations higher than 1000 pCi/l may relate to these sources which may be the uranium anomalies in the underlying formations.

The results of the natural thermoluminescence studies of the overburden sand formations makes it possible to distinguish between several modes in the statistical distribution of the TI intensities (Fig. 6). In comparison with the "normal values" of TI it is

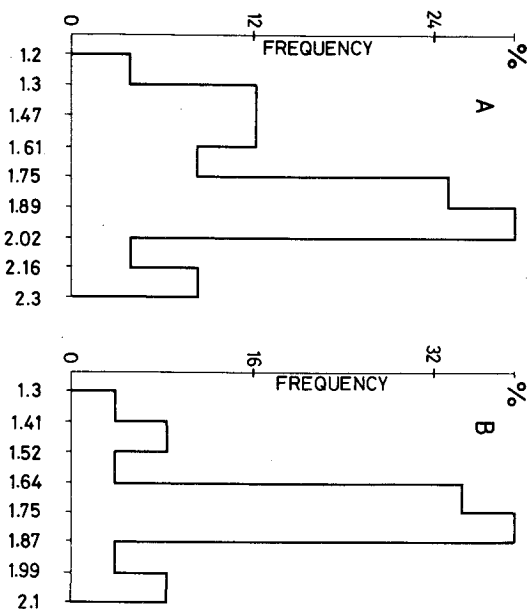


Figure 6

Frequency histograms of the TI intensities (H_{2n}), third period of field work, at three meters depth, results for the whole rock (A) and for quartz (B).

possible to define negative and positive anomalies. The phenomenon and origin of the negative anomalies have been discussed in a previous paper (Charlet, Dupuis, Quinif, 1979). They are related to a reduction of the thermoluminescence signal in the superficial zone of the sand formation as the result of brushwood fires or exposure to sunlight (Wintle, 1981) followed by the movement of the sediments from the top to the bottom of the hill (a rehandling phenomenon with a bimodal granulometric distribution). The negative TI anomalies can often be avoided by taking samples with a screw-auger at a depth of a few meters.

The results of the second and third periods of field work showed that the positive anomalies are located in the central and southern part of the profile centred on the points 16 and 28-30. This confirmed the results obtained in 1978. The definition of the anomalies is improved by increasing the depth of sampling (Fig. 8) and by choosing a mineral of high TI characteristics for analysis (Fig. 7). In addition these anomalies (Fig. 8) are nearly located directly above the uraniferous occurrences detected in the underlying formations by the radiometric survey but are a little shifted in comparison to the radon anomalies detected by using the other integrated methods. It should be noted that the integration times are much greater for natural thermoluminescence than for the integrated radon detection methods used in field work.

The "normal" TI values have been described by using the mean of the values in a experimental error around the maxima of the principal mode. The values calculated in a similar manner for the H_2 and for the glow curve shape parameter R are given in the following table (Tab. 2) which are the results of the third period of field work.

Table 2.- Mean values for the TI parameters, third period of field work.

| Depth | | 0,5 m | 2 m | 3 m |
|---|----------|-------|-------|-------|
| The whole rock | H_{2n} | 1,326 | 1,734 | 1,825 |
| | R | 49,3 | 50,04 | 50,05 |
| Quartz | H_{2n} | 1,595 | 1,717 | 1,753 |
| | R | 29,4 | 30,6 | 29,7 |
| Radon concentration (calculated in pCi/l) | | 251 | 658 | 777 |

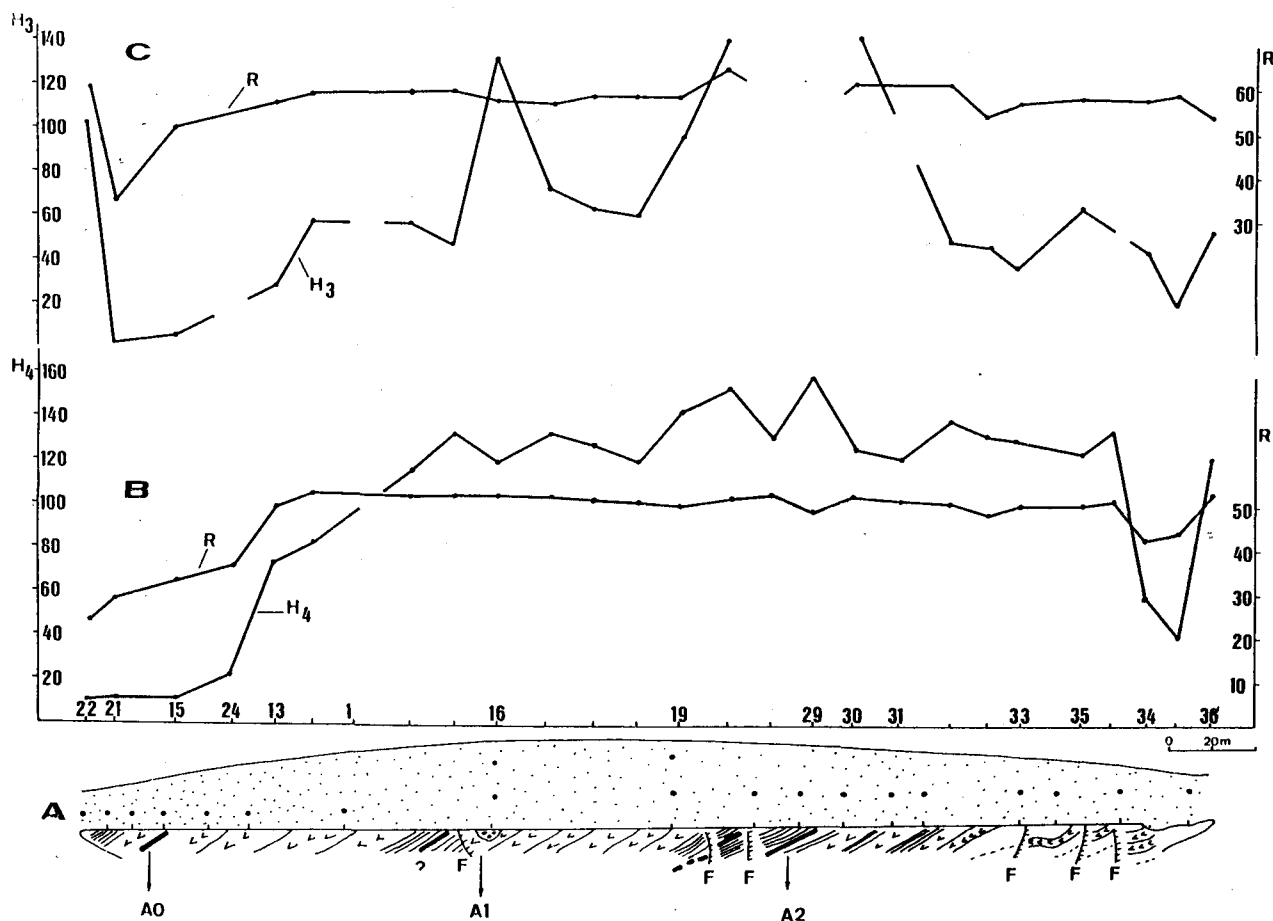


Figure 7

Natural thermoluminescence results for K feldspar (in C) and for quartz and the plagioclases (in B), sampling at about 0,5 meter depth.

It can be seen that the glow curve shape parameter R does not vary with the depth of sampling, this is related to the presence of the same-feldspathic material in all cases. The TI intensity level (H_2n) increases with sampling depth. In addition the relative increase with depth is greater for the whole rock than for the quartz itself. This table also gives the radon concentration calculated from the emanation theory (Alekseev *et al.*, 1959) and by using the radium distribution measured in the Landenian sands by γ -ray spectrometry. It appears that the "normal" values of the TI intensity follow a similar dependency as the radon concentration in the overburden sand formations.

CONCLUSIONS

The three integrating techniques of radon detec-

tion used in this work all showed general agreement (Fig. 5) in the locating of the principal radon maxima and minima in the overburden sands at Blaton. The general agreement obtained should not be taken to mean that all the techniques are equally sensitive and useful under all geological and climatological field conditions. In order to emphasize this point a short resume is given below of the advantages and disadvantages of the four techniques.

NATURAL THERMOLUMINESCENCE OF THE OVERBURDEN FORMATIONS

It is very different from methods of radon detection proposed until now as it uses natural detectors formed by the minerals of the overburden formations. The integrating time can thus be very long (some millions of years) and a fossil effect can be observed in a

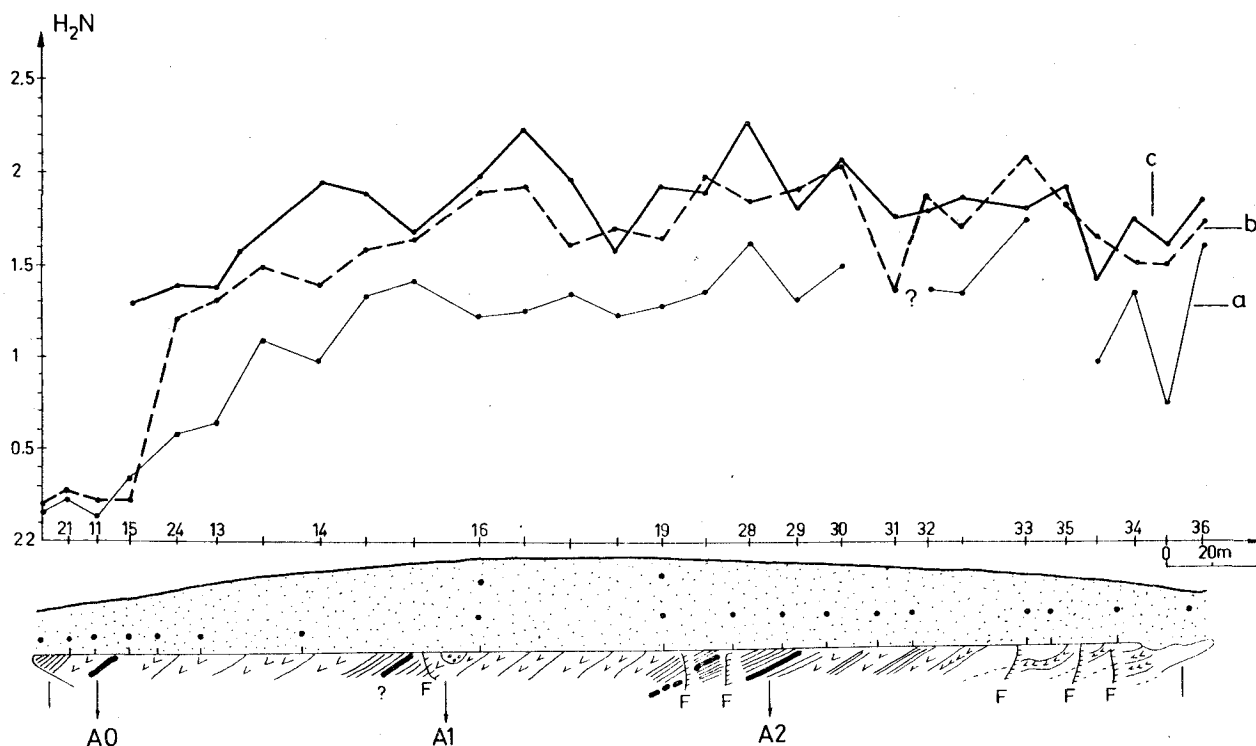


Figure 8

Natural thermoluminescence results for the whole rock, third period of field work, sampling at a) $\pm 0,5$ meter ; b) two meters ; c) three meters.

zone where there is at present no radon migration (permanent wet climatic conditions, etc.).

With this technique only one visit to the field is necessary while the others radon techniques, two field visits are necessary for the setting and the recovery of the detectors. However several natural disturbing factors must be eliminated : the rehandling phenomenon with a reduction of thermoluminescence, sometimes the sedimentological effects in relation with a variation of the origin of the detrital material (variation of the features of the natural detector). Besides one often has perform laboratory operations for extracting the various mineralogical phases (quartz, feldspars, ...).

TLD (THERMOLUMINESCENCE DOSIMETRY)

It is a classic integrated method with the same general characteristics as the alpha track or charcoal techniques. With this technique two visits to the field are necessary. Like the alpha track method the integrated time can be longer than with the techniques based on the equilibrium between radon and its radioactive

daughters (charcoal, alphacard, ...) but its advantages in regard to the alpha track methods is the very short delay in obtaining the results. Compared however with the charcoal it is more difficult to distinguish the background effect and the technique would have to improve by a selection of the more sensitive Tl powders.

For the F.P.Ms. laboratory the advantage of this system results in the utilization of the same reader as the one used in natural Tl studies.

CHARCOAL TECHNIQUE (C.A.R.P.)

The method is simple and inexpensive to operate, does not require skilled personnel and can distinguish between the Radon-222 signal and any Thoron noise present. The data may be rapidly and confidentially obtained directly by the operator in the field if necessary. It is very sensitive and even with a simple shielded scintillation detector in the field it can easily detect radon levels of 100 pCi/l which is typical of near surface background or supported radon concentrations in normal soil. It is, however, a restricted integrating method with an effective memory of approximately

two weeks. Its efficiency is severely reduced in wet or high temperature environments and is best suited to cold dry conditions. It is to be noted that under these conditions the effectiveness is also reduced of methods based on the use of alpha track plastics.

ALPHA TRACK TECHNIQUE

This technique which is simple to use gives an alpha signal integrated over the full period of burial. The alpha tracks obtained may not be exclusively due to radon and its daughters. A diffusion barrier will, however, eliminate thoron interference. Because the microphysics of the alpha emitters in the air cavity of the probe is not fully understood it is not possible to convert from alpha track densities to mean radon concentration. Because the alpha track plastics must be etched and counted there is often a considerable delay between probe recovery and obtaining the data. In this work the plastic was etched and the alpha tracks were counted by the investigators.

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