

A CLOSER LOOK AT THE NATURAL RADIOACTIVITY IN SOILS

by: H.Surbeck and G.Piller
Lab.Fribourg of the Swiss Federal
Commission for Radioactivity
Surveillance (KUEr)
Perolles
CH-1700 Fribourg, Switzerland

ABSTRACT

Enhanced indoor radon levels are frequently found in the western part of the Swiss Jura mountains, a karst region. The ^{226}Ra activity in the limestone bedrock is low (20 Bq/kg). In the thin soil overburden the ^{226}Ra activity concentration is 2 to 3 times higher than in soil samples from the Swiss Plateau but on the average still below 100 Bq/kg. It is supposed that the radon problem in this region is due to the high permeability of the karst bedrock.

A sampling device for radon in soil gas measurement is described that permits a coarse in-situ soil permeability determination.

Specific activity vs. grain size measurements for some Swiss soils and the high activities found in heavy minerals are presented and a possible aeolian origin of the soils in the Western Jura is discussed.

INTRODUCTION

Predictions of high radon risk areas based on "typical" U-concentrations of the bedrock generally show a poor correlation with measured indoor radon concentrations (1,2). Better correlations are achievable using soil maps (1), showing the dominant role of the soil properties, the uranium in the bedrock being of less importance (3).

In Switzerland there are places too where geologic maps of the bedrock fail to predict high indoor radon concentrations. In the Rhone valley where several uranium mineralizations are known (4) no high indoor radon concentrations are measured, whereas in the Western Jura mountains, consisting mainly of limestone, houses with up to 5kBq/m^3 of radon in living rooms are found.

Apart from some rare exceptions, covering some tens of km^2 , there are no soil maps in Switzerland. This may be due to the geologist's preference for solid rocks, neglecting the overlaying mud. But this mud we are living on may have a nuclide composition and nuclide concentrations very different from the underlying bedrock.

As an example we mention the gamma dose rate measurements carried out 1961 in Switzerland (5). The terrestrial component turned out to be low on limestone, high on the crystalline rocks of the Alps and intermediate on Tertiary sediment (often overlaid by Quarternary deposits). These measurements were occasionally cited to exclude any natural reason for the enhanced radon concentrations observed in the Western Jura. A look at the original data shows a clear preference of the authors to take measurements in quarries and at other sites where the bare bedrock was visible. Results from recent in-situ gamma spectrometry measurements (6) are in clear contrast to this picture of a low activity Jura. The ^{226}Ra activity concentrations in soils of the Western Jura are found to be among the highest in Switzerland.

We don't deny the usefulness of geological maps for the search for high radon risk areas. But we want to show that a closer look at the soil overburden helps to understand why predictions based on geologic maps may fail.

If the soil overburden is very thin, as is frequently the case in the Jura mountains, the first few meters of the bedrock

deserve a closer look too.

Domains other than the radon research may also profit from a closer look at the natural radioactivity in soils. As an example we mention the use of natural heavy mineral placer deposits (having high specific activity) to study the sand transport in dunes (7,8).

In this paper we present our experimental methods used to study soils and the application of these methods to reveal the origin of the radon problem in a part of the Jura mountains.

THE SITE UNDER STUDY

High indoor radon concentrations (up to 5 kBq/m³ in living rooms) are found in a city lying in the western part of the Swiss Jura mountains at 1000 m above sea-level (see figure 1). The city is well-known for its watch industry and therefore there are clear ²²⁶Ra contaminations in and around some former radium-processing workshops. But high radon concentrations are also found in houses with no radium contamination and lying far away from these workshops (9). In addition there are several former radium processing workshops in other parts of Switzerland where enhanced radon concentrations are limited to the still slightly contaminated workshops and even adjacent houses show no enhanced radon concentrations. So there are clear indications that at least an important part of the radon problem in this city has a natural origin.

At one place in this city a cluster of houses with high indoor radon concentrations has been found. Winter averages of the ²²²Rn concentrations are shown in figure 2. Concentrations are two to three times higher in winter than in summer. In one of the houses shown in figure 2 (AR) ²²⁶Ra-activated luminous paint was processed until 1963. Small radium contaminations are still present in and around this house but the total activity of the contaminations located so far is at least an order of magnitude too small to explain the radon concentrations in this house. The other houses are free from radium contaminations.

The bedrock at this site consists of Jurassic limestone (Malm) with a thickness of at least 400 m. The Malm in this region frequently forms a karst. The porosity of the undisturbed Malm-limestone is very low, not exceeding 0.5% (10). The high but heterogeneous permeability of this karst formation is due to a network of channels with a large range of

diameters. Water flows through this network with velocities from 5 to 300 m/h (11). The largest channels form caves that can be visited. One of these larger underground cavities is about 200 m from the houses shown in figure 2. The soil overburden at this site has a thickness of 20-80 cm.

ACTIVITIES OF THE U- AND TH-SERIES IN BEDROCK AND SOIL

An extensive literature exists on the U and Th concentrations in rocks. An important part of the published data is based on measurements of the radon decay products with NaJ-detectors assuming a perfect equilibrium all along the U- and Th-series, calling the radon decay products that remained in the sample "U" or "Th". An equilibrium down to the ^{226}Ra or ^{224}Ra may exist in igneous rocks and old sediments. But in recent sediments and soils an equilibrium is rather the exception than the rule (12). There is also the problem of the radon escape from the sample. Due to the inert nature of the radon and the large concentration gradient even a small crack or gap in the sample container may lead to an important radon loss if the sample is not completely dry (13).

EXPERIMENTAL METHODS

Normally soil samples are taken to a depth of 10 cm and split into two parts (0-5 cm and 5-10 cm respectively). At places where soil gas measurements are carried out soil samples are also taken at a greater depth. The grass is cut away but the roots are left. At the laboratory the soil samples are dried under an IR-lamp.

For each type of soil encountered the grain size distribution is determined allowing a coarse geologic characterization of the soil. The procedure to determine the grain size distribution is described below.

Rock samples are only slightly crushed to fit into the sample containers.

The activities of the radionuclides are determined by gamma spectrometry using a Ge(Li)- or a HP-Ge-detector. For routine work 500 to 1000 cm³ of the soil or rock samples are measured immediately after drying. For samples that large and measuring times of one day the ^{226}Ra activity can be determined directly, there is no need to wait for the buildup of the

radon.

For small samples (up to 40 cm³) a different technique is used. The sample is placed in a flat thin-walled polystyrene box, saturated with water, deep-frozen and kept for at least 3 weeks in the deep-freezer prior to measurement to allow for the buildup of the radon. A thermoelectric cooler keeps the sample frozen during the measurement (one day). The ice turned out to be a very efficient barrier against radon diffusion. Compared with other seals like epoxy resins, ice has the advantage that after thawing and drying no trace of the seal is left on the sample. Another advantage of the ice as seal is described below.

RESULTS

²²⁶Ra and ²²⁸Ac activity concentrations for some Swiss soils are shown in figure 3. "Weighted" frequency in figure 3 means that every data point is represented by a gaussian with unit area and a width corresponding to the statistical error. The partial areas of each gaussian, falling into one class, are summed up to give the "weighted" frequency in this class.

Whereas the ²²⁶Ra values for the samples from the Swiss Plateau are centred around 35 Bq/kg, samples from the Western Jura show a second peak in the frequency distribution at about 75 Bq/kg. Samples from places where there were indications of a contamination by industrial activities are not included in figure 3. For the ²²⁸Ac activity concentration there is no significant difference between the two sample sets.

In 6 samples from the limestone bedrock at the site under study (21 +/- 2) Bq/kg ²²⁶Ra and (2 +/- 0.5) Bq/kg ²²⁸Ac were measured.

FRACTIONATED SAMPLES

To know where the radionuclides are located in a soil may help to reveal its origin and history. For the radon research it is of particular interest to know where the ²²⁶Ra is sitting. Inspired by the work of Megumi et al.(14) and de Meijer et al. (7,8) we started to measure the specific radionuclide activities vs. grain size in Swiss soils (13) and the radionuclide concentrations in heavy mineral fractions.

EXPERIMENTAL METHODS

The dried soil samples are sieved using stainless steel sieves with mesh widths of 5mm, 2mm, 1mm, 0.5mm, 0.25mm, 0.125mm and 63 μ m. The <63 μ m fraction is separated by dry sieving. For loamy soils more than 70% of this fraction stays sticking to the larger grains. The larger grain size fractions are therefore separated by wet sieving and dried. The quantity of the water used is kept as low as possible and corresponds to about the amount of rainwater that has passed naturally through the soil during the last 10 years. The water used is ordinary tap water with a pH slightly below 7. To get enough material in every fraction we need about 500 g of the dried soil. This quantity can be treated in one batch.

The heavy mineral fractions were collected by an experienced gold prospector (15) using a gold pan ("batea").

The samples are measured using the deep-freeze method. Before we changed to the deep-freeze method the well dried samples were filled into the same type of polystyrene boxes as used for the newer method and the boxes were sealed by gluing. This method works well if the sample is kept completely dry for the radon emanation from dry samples is low .

The deep-freeze method opens an easy way to determine what part of the radon produced is able to leave the grains. After measuring the frozen sample the thermoelectric cooler is switched off and the sample is allowed to thaw. The radon decay products measured in the thawed sample stem from radon atoms that cannot leave the grains. Due to the small distance to be traversed in the water and the leaks in the sample container radon atoms that have entered the surrounding water are lost. This additional measurement is not yet used in routine work. We just verified that no appreciable amount of radon is retained by the water.

RESULTS

Specific activities vs. grain size for three soil samples from the site described above and for two samples from the Swiss Plateau are shown in figure 4. The latter samples show a clear increase of the activity concentration with decreasing grain size. This behaviour we expect for a soil of fluvioglacial origin . In the samples from the Western Jura the activity in the silt and clay fraction (<63 μ m) is

significantly lower than in the 63-125 μm fraction.

Figure 5 shows that very high specific activities can be found in heavy minerals.

SOIL GAS

Even a soil with an enhanced radon production will not create a radon problem if the soil permeability is low. On the other hand, high permeabilities ($> 10^{-11}\text{m}^2$) alone may cause elevated indoor radon levels (16). Therefore it is not sufficient to determine the radon concentration in the soil gas. Some sort of in-situ permeability determination has to be made too.

EXPERIMENTAL METHODS

The apparatus used for soil gas measurements shown in figure 6 is similar to the one described by Tanner (17). Approximately 1000 cm^3 of the soil gas are pumped generally from a depth of 50 cm, stored for 5 min in the pump to allow for the ^{220}Rn to decay and are then transferred through a filter retaining the aerosols to a Lucas-cell. After the transfer of the soil gas to the Lucas-cell we wait for 5 min and then count the alpha decays for 10 min.

To gain information on the permeability of the soil we refill the pump with outside air and measure the time needed to pump at constant pressure this air volume into the probe hole. The constant pressure is produced by the weight of the pump piston. The time needed to press 1000 cm^3 at a pressure of 4 kPa into the hole is on the order of 10 sec for a soil permeability of 10^{-12}m^2 .

The radon concentration measured is divided by the time needed to pump the outside air into the borehole (after subtraction of the pump, valve and tubing contribution). This number takes into account not only the radon concentration measured but also the soil permeability and the unknown leak around the packer. It's a better estimate for the "radon availability" than the bare radon concentration.

The whole equipment can be transported and operated by a single person and is fully battery operated. With one equipment we can measure up to 20 points a day.

Tests on the reproducibility of the measurements are promising. With the packer only partly inflated the measured radon concentrations are up to 10 times lower than the values obtained with the packer fully inflated, but the "normalized" values vary by less than 20%.

Having recognized the importance of the soil permeability and the unavoidable leaks we will wait to present any of our radon in soil gas results until they are confirmed by measurements with the new sampling device.

In a cave near the site under study we have measured 20 kBq/m³ in the air at a depth of 6 m and 40 kBq/m³ at 25 m below ground.

At the moment we don't know how to get reliable results for the permeability of the karst bedrock. The only (water) permeability data available for the Western Jura are from pumping tests in deep boreholes (10). But the author states that the data are not representative due to the large inhomogeneities found in a karst bedrock.

DISCUSSION

Figure 3 shows that the ²²⁶Ra activities in soils from the Western Jura are generally 2 to 3 times higher than in soils from the Swiss Plateau. This fact has recently been confirmed by in-situ gamma spectrometry measurements.

But even a ²²⁶Ra activity of 100 Bq/kg does not explain the high indoor radon levels frequently found in the Western Jura and until now no other important radon source has been found. The limestone bedrock is a rather poor radon source, containing only about 20 Bq/kg of ²²⁶Ra. Two clay samples from underground cavities (2m and 25m below ground) showed ²²⁶Ra and ²²⁸Ac activities similar to the ones found in the soil.

We suppose that the radon problem in this region is due to the high permeability of the karst bedrock. It's numerous interconnected channels form an efficient gas transport system. Radon degassing from percolating water that has been saturated with radon during its passage through the soil may feed this underground radon distribution system.

The origin of the soil found at the site under study is unclear. According to Pochon (18) this type of soil, frequently found in the Western Jura, is not derived from the

weathered bedrock but from the weathering of wind-borne (aeolian) material deposited at the end of the latest glacial period (Würm). The moraines of the retreating Rhonegletscher are a probable origin for the aeolian dust. This claim is supported by the fact that an important concentration of heavy minerals has been found in soils from the Western Jura (19), in particular a mineral association characteristic for the schists of Casanna (Rhone valley). This presence of heavy minerals from the Rhone valley, having high specific activities (figure 5), may explain an enhanced ^{226}Ra concentrations in soils from the Western Jura, but the $^{226}\text{Ra}/^{228}\text{Ac}$ ratios in these soils are larger than the ratios found in the heavy mineral samples.

Transport by wind clearly modifies the grain size distribution and may lead to an enrichment of heavy minerals in the smaller grain size fractions, leading also to higher specific activities in these fractions. It remains to be proved that the activity vs. grain size pattern observed in soil samples from the Western Jura (figure 4) is really due to the presence of aeolian material.

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.

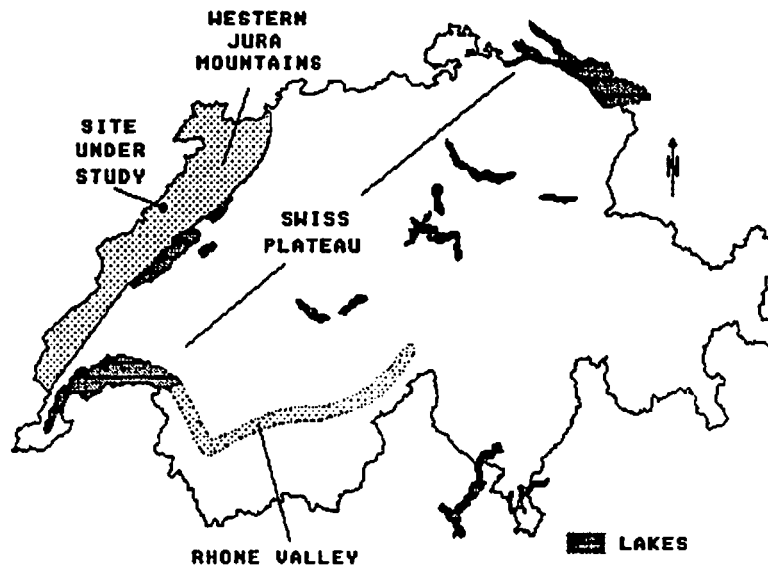


Figure 1: Map of Switzerland.

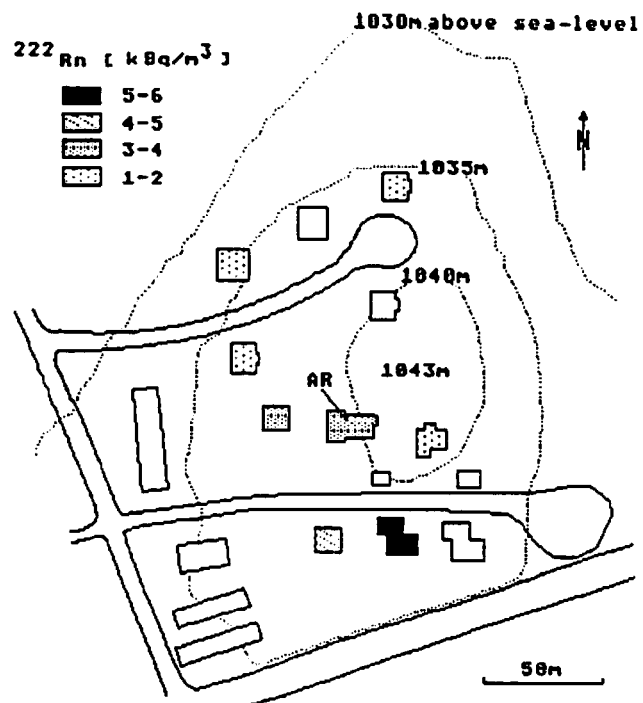


Figure 2: The site under study. AR = former radium processing workshop.

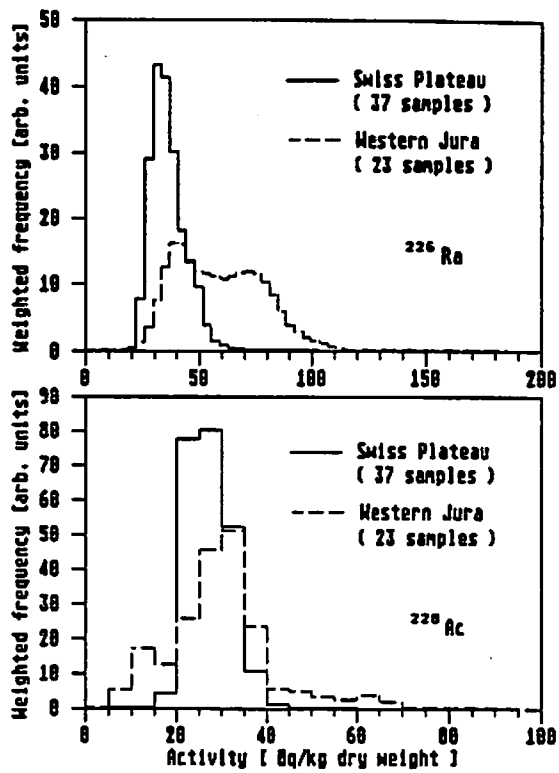


Figure 3: Activities in Swiss soils. The term "weighted frequency" is explained in the text.

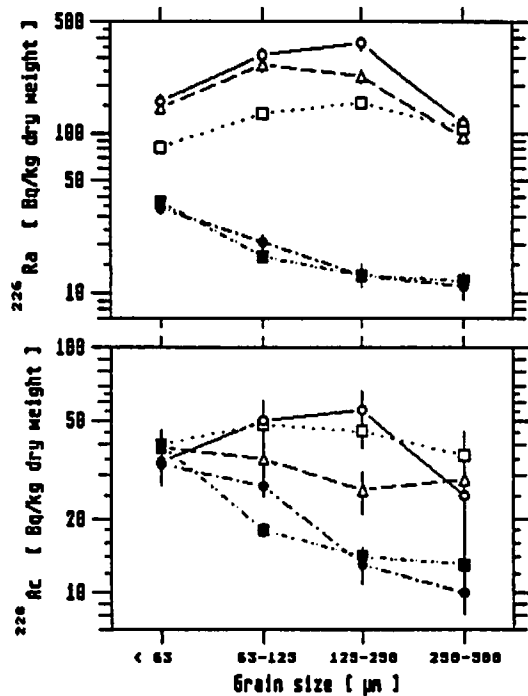


Figure 4: Activities vs. grain size for soils from the Western Jura (open symb.) and Swiss Plateau (filled symb.).

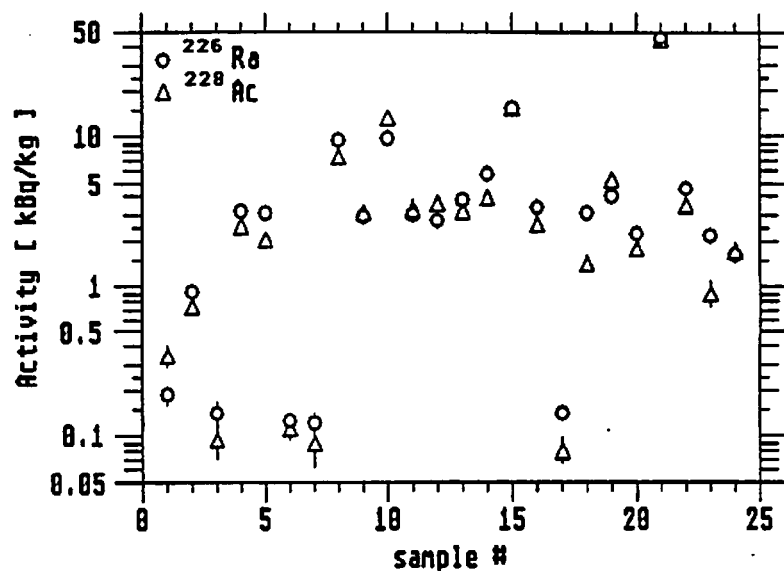


Figure 5: Activity concentrations for some heavy mineral samples separated from natural placer deposits in the Rhone valley (upstream Lake Geneva).

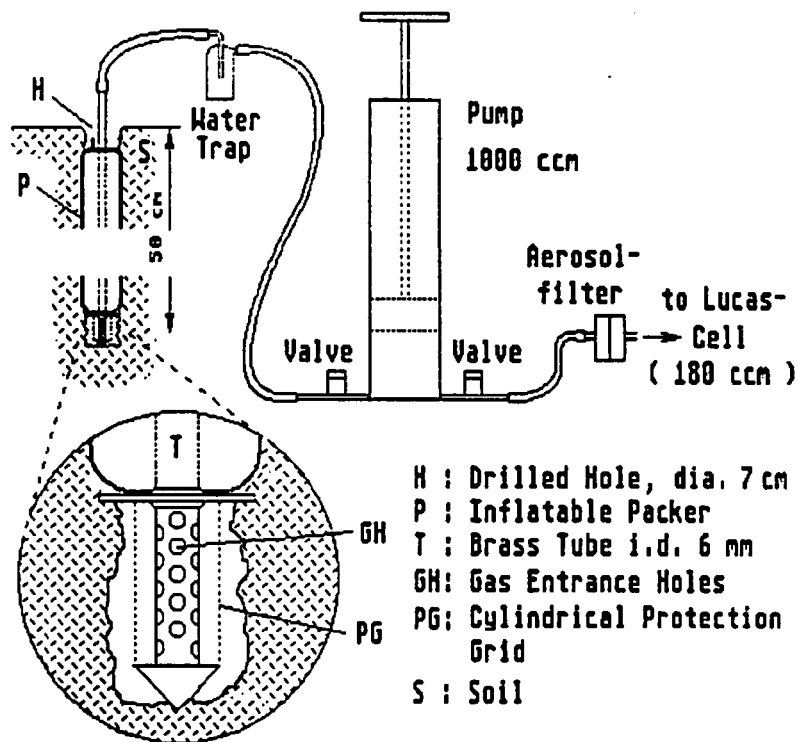


Figure 6: The equipment used to sample soilgas and to measure soil permeability.

REFERENCES

- 1) Castren, O., Voutilainen, A., Winqvist, K. and Mäkeläinen, I. Studies of high indoor radon areas in Finland. The Science of the Total Environment, 45 (1985) 311-318.
- 2) Damkjaer, A. and Korsbech, U. An investigation of the possible correlation between the local geology and the indoor radon concentration. Paper presented at the Fourth International Symposium on the Natural Radiation Environment, Lisboa, Portugal. December 7-11, 1987.
- 3) Revzan, K.L., Nero, A.V. et al. Mapping surficial radium content of U.S. soils as a partial indicator of indoor radon concentrations. Paper presented at the Fourth International Symposium on the Natural Radiation Environment, Lisboa, Portugal. December 7-11, 1987.
- 4) Labhart, T.P. and Rybach, L. Granite und Uranvererzungen in den Schweizer Alpen. Geol.Rdschau 63 (1974) 135-147.
- 5) Halm, Ed., Herbst, W. and Mastrocola, A. Messung des natürlichen Strahlenpegels in der Schweiz. Beilage B zum Bulletin des Eidg.Gesundheitsamtes Nr.6 (1962).
- 6) Jahresbericht 1985/86 der Eidg.Kommission zur Ueberwachung der Radioaktivität. In press.
- 7) De Meijer, R.J., Put, L.W., Bergman, R., Landwehr, G., Rizebos, H.J., Schuling, R.D., Scholten, M.J., Veldhuizen, A. Local Variations of the outdoor radon concentration in the Netherlands and physical properties of sands with enhanced natural radioactivity. The Science of the Total Environment 45 (1985) 101-109.
- 8) De Meijer, R.J., Put, L.W., Schuling, R.D., De Reus, J., Wiersma J. Natural radioactivity of heavy mineral sands: a tool for coastal sedimentology? In: Proceedings of the International Symposium on Radioactivity and Oceanography. Cherbourg, France, June 1987.
- 9) Surbeck, H. The search for radon sources, a multidisciplinary task. Paper presented at the Fourth International Symposium on the Natural Radiation Environment, Lisboa, Portugal. December 7-11, 1987.

- 10) Tripet, J.P. Etude hydrogeologique du bassin de la source de l'Areuse. Thesis, University of Neuchatel, Switzerland, 1972.
- 11) Burger, A. Hydrogeologie du bassin de l'Areuse. Bull.Soc.neuch.Geogr. 52 (1959) 304
- 12) Gascoyne, M. Geochemistry of the actinides and their daughters. In: Ivanovich, M. and Harmon, R.S. (eds.), Uranium Series Disequilibrium. Claredon Press, 1982, p.53.
- 13) Surbeck, H. and Völkle, H. Radionuclide content vs. grain size in soil samples. The Science of the Total Environment 69 (1988) 379-389.
- 14) Megumi, K. and Mamuro, T. Emanation and exhalation of radon and thoron gases from soil particles. J.Geophys.Res. 79/23 (1974) 3357-3360.
- 15) Mäder, F. Goldspuren in den Sanden einiger Bäche des Einzugsgebietes der Broye (Kt.Freiburg). Bull. Soc.Frib.Sc.Nat. 72 1/2 (1983) 56-74.
- 16) Peake, T.R. Radon and geology in the United States. Paper presented at the Fourth International Symposium on the Natural Radiation Environment, Lisboa, Portugal. December 7-11, 1987.
- 17) Tanner, A.B. A tentative protocol for the measurement of radon and thoron availability from the ground. Paper presented at the Fourth International Symposium on the Natural Radiation Environment, Lisboa, Portugal. December 7-11, 1987.
- 18) Pochon, M. Origine et evolution des sols du Haut-Jura suisse. Memoires de la Societe Helvetique des Sciences Naturelles. Vol.XC. 1976
- 19) Matthey, F. Contribution a l'etude de l'evolution Tardie et postglaciaire de la vegetation dans le Jura Central. Mat. pour le leve geobot. de la Suisse. 53 (1971) 86.