



Radon and Space Radiation Protection Measurements

Ph.D. dissertation

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Jelölt

Tanúsítom, hogy Dr. Csige István doktorjelölt 1995-1997 között a fent megnevezett doktori alprogram keretében irányításommal végezte munkáját. Az értekezésben foglaltak a jelölt önálló munkáján alapulnak, az eredményekhez önálló alkotó tevékenységével meghatározóan hozzájárult. Az értekezés elfogadását javaslom.

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Dr. Hunyadi Ilona
Témavezető

Preface

I have been working with etched track detectors since the end of 1984. At that time Dr. György Somogyi, who was one of the highest respected scientists of the world in the field of solid state nuclear track detectors, invited me to his laboratory in the Institute of Nuclear Research (Atomki) of the Hungarian Academy of Sciences (Debrecen, Hungary) to carry out experimental studies on cosmic ray charged particles. After receiving my M.Sc. degree in physics in 1986, I became employed in the laboratory as a research assistant. The Hungarian Intercosmos Council granted my position. We continued working on space radiation dosimetric measurements in collaboration with partners from the Institute of Biomedical Problems (Moscow, Russia [formerly Soviet Union]). In the spring of 1987 Dr. György Somogyi suddenly died of heart attack. After his death, Dr. Ilona Hunyadi took the lead of the group.

Gradually, but still parallel with space radiation measurements, I became interested and involved in environmental alpha radioactivity studies and especially in radon dosimetric measurements. Soon, in 1989, I became the leader of a project, which aimed at developing a small volume, passive, etched track type radon monitor and an automatic image analyzer to count tracks. We worked on this project in collaboration with the Mechanical and Optical Corporation of the Hungarian Optical Works (Mátészalka, Hungary). The project was successfully finished in 1990. At the end of 1990, I have submitted my thesis to obtain my *dr. univ.* degree. The title of the thesis was “Development of solid state nuclear track detector technique and its application in space and radon dosimetry”. I have defended my thesis and have obtained the *dr. univ.* degree of the Lajos Kossuth University (Debrecen, Hungary) in 1991.

In the years of 1991 and 1992, following an invitation from Prof. Dr. Eugene V. Benton (who is another one of the greatest “track scientists” of the world), I worked at the Physics Research Laboratory of the University of San Francisco (California, United States) as an exchange visitor. There we have measured cosmic ray charged particles that were recorded in solid state nuclear track detector stacks, which stacks were exposed to space environment for almost 6 years on the so-called Long Duration Exposure Facility satellite. This work aimed at gathering information on space radiation environment in a low Earth orbit, an orbit onto which the space station “Freedom” was planned to be launched in those years. The National Aeronautics and Space Administration (NASA) of the United States has supported our work.

At the beginning of 1993, after 2 years in the United States, I have decided to return to Debrecen, Hungary. I have received a full time permanent position in the Institute of Nuclear Research and joined again the Track Detector Group. There, I have decided to focus my studies on radon research. First, I have got involved in some of the group’s ongoing projects, which were led by Dr. Ilona Hunyadi. Those projects aimed to study radon in the environment, especially the behavior of radon in caves and radon related phenomena at a peculiar radon spot area in Hungary (at Mátraderecske village in the Mátra Mountain, where enhanced CO₂ geo-gas up-flow carries radon into houses).

Later on I, too, became leader of several projects. These projects aimed at studying radon transport in porous media and doing radon concentration measurements mostly for dosimetric purposes. In the summer of 1994, a year after Dr. Ilona Hunyadi has formally retired; I have

become the leader of the group. At that time we have also changed the name of the group, which is now known as the Radon Group of the Atomki (Debrecen, Hungary). Since the summer of 1996, Dr. József Hakl leads the group.

During this about 10 years scientific carrier of mine, I have attended about 50 scientific meetings, co-authored 47 scientific papers and delivered about 80 scientific lectures. I have referred about 50 papers and co-edited a special issue of the Journal of Radiation Measurements.

Currently, we are working on several radon related projects, which are supported mostly by the Hungarian National Scientific Research Fund. In one of these projects, which I am the leader of, we aimed at developing a procedure to determine radon source potential of building sites. This work is based on study of radon transport in soil and its entry into houses at selected “radon test-houses”. We work on this project in international collaboration with partners from different European Union countries, including France, Germany, Sweden and Spain. After a few years in the “radon business” I am convinced that, though much work have been done, but still, there is a lot more to do. I believe, that our efforts in the Radon Group of Atomki are right and worth things to do.

Finally, I should like to say many thanks here to my colleagues, with whom I enjoined to share the fascinating works we made together (both in the fields and in the labs) and then the excitements learning some small secrets of nature. These closest colleagues are Dr. József Hakl, Dr. Ilona Hunyadi, Attila Vásárhelyi, Prof. Dr. Eugene V. Benton, Eric R. Benton and Luis A. Frigo.

Also, I should like to thank many other colleagues and collaborators, with whom I had the good luck to work together. Some of these are Dr. István Lakatos, Dr. András Várhegyi, Gábor Géczy, Dr. László Lénárt, Dr. István Töröcsik, Dr. K. Oda, Dr. M. Fujii, Dr. A. M. Marenny, Dr. G. P. Guertzen, Dr. N. Nefedov, and Dr. R. Henke.

Particularly, I should like to thank to Mrs. Enikő Molnár for the excellent job and help she has done for us in the Radon laboratory of Debrecen.

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1. Introduction

In this dissertation I will present some of the results of the scientific research work that I have done in the fields of cosmic ray charged particle and environmental radon dosimetric measurements during the past decade. My aim with this dissertation is to obtain the Ph.D. degree in physics of the Lajos Kossuth University (Debrecen, Hungary).

This dissertation deals with radiation protection. To be more specific the first part deals with protection of the crew of manned space flights against the exposure to cosmic ray charged particle irradiation in low Earth orbit. The second part deals with protection of the general population against exposure to inhaled radon and its decay products present in air in the environment and particularly in homes and in workplaces. Though these two fields may seem very different applications of radiation protection, the thing that brings them together in this dissertation is that we have used the same detector type, i.e. etched track detector based techniques, in most of the measurements in both of these two fields. Other techniques were also used, but here I will focus on measurements, which we did with etched track detectors. That is because I think that my part in or contribution to these “track works” was always crucial and was decisive in most of the cases.

From our natural radiation environment our bodies are constantly bombarded by different kinds of radiation. Some of these radiations are useful or even crucial for our life on Earth (like those coming from the Sun). In another aspect some radiations seem to be very friendly (like first strokes of springtime Sunshine), while others (mostly ionizing radiations) are known to be dangerous. There are serious opinions, however, (though not generally accepted) that even in the case of ionizing radiation small doses might do more good, than harm. Little is known about the health effects of low doses. It is absolutely sure, however, that higher doses of ionizing radiation cause almost always more detriment, than do anything good.

Taking unnecessary, unjustified extra risk is against our evolutionary human nature. Risk and cost versus benefit estimates are part of our every day life. Radiation risks, especially those of low doses, however, are poorly known even for experts. The fear of the unknown pushes us to learn, to think, to decide and to act.

According to the International Commission on Radiological Protection (ICRP) an effective dose of 1 mSv corresponds to a risk (or probability) of about 5.6×10^{-5} (for workers) and 7.3×10^{-5} (for the general public) of detriment caused by ionizing radiation (ICRP-60, 1990). The detriment is a weight sum of different damage the radiation might cause, including fatal and non-fatal cancer, hereditary effects and the length of life lost or impaired.

Crewmembers of spacecrafts on low earth orbit receive effective dose around 1 mSv/day. Dose rate may vary significantly depending on orbit parameters, and may also depend on time. Much of the dose is received when the space vehicle crosses the so-called South Atlantic Anomaly. Protons, trapped in the Van Allen radiation belts contribute to the dose dominantly. Energetic solar flare particles may dominate the total dose during active Sun periods. In our experiments on the so-called Long Duration Exposure Facility satellite, we have studied the flux and the linear energy transfer (LET) and directional distribution of cosmic ray charged

particles. From our studies we have learned that the high linear energy transfer portion of the measured LET spectrum is some two to three order of magnitude higher than previously estimated and that it is made up mainly by secondary particles generated in collisions of high energy protons with the nuclei of the detector material. Our measurements have confirmed the flux and directionality of the trapped protons and indicated some anisotropy of galactic cosmic rays. These results will be discussed in this dissertation.

Inhalation of short-lived radon decay products, present in air of homes and of workplaces, causes an average effective dose of about 1 mSv/year for a person from the general population. That is about half of the effective dose people receive in a year from exposure to the natural radiation environment. The principal detriment due to the inhalation of radon and its progeny is that associated with the fatal lung cancer. The risk factor of exposure to high doses of radon is relatively well known from studies of some underground miner groups. Epidemiological surveys among the general population recently confirmed similarly increasing risk for those living in houses with such elevated radon levels that the lifetime exposure of inhabitants becomes comparable to those in miner studies. Results of studies at normal levels of indoor radon concentrations are more controversial or often difficult to analyze.

Nevertheless, indoor radon surveys showed that houses with high radon concentration could be found with considerable probability. Finding these houses, and reducing radon in them is the motivation of many efforts and was one of the indirect scopes of some of our works, too. In the Institute of Nuclear Research we have developed methods, based on etched track and semiconductor detectors to measure radon concentration in-situ in indoor air, in soil gas and in natural waters. We have measured radon concentration primarily for radon dosimetric purpose in the air of many houses and in some underground workplaces (mostly in caves). We have found homes and workplaces as well, where radon concentration was higher than that could be conveniently accepted. Based on our measurements, we have suggested responsible local and national authorities to take mitigation, and in some cases regulatory actions.

In a more general approach, efficient search strategies for radon-prone areas and efficient mitigation techniques require deep level of understanding of radon occurrence in nature, its origin, its transport in soil, and its entry into houses. Influencing geologic factors, soil properties, weather parameters, house characteristics and inhabitant behaviors make the problem very complex and call for collaboration of experts from all relevant fields. The scientific aim of one of our recent work is to develop criteria for building sites and for indoor radon concentration, when all relevant influencing factors are taken into account. This work is in progress, however, I decided to show some early results mainly to indicate what are the recent problems that scientists in “radon business” face today, and what the promising scenarios might be that show towards a safer, healthier natural radiation environment.

2. Literature Overview

2.1. Space Radiation Protection

Space exploration is one of the most exciting areas of today's scientific research. Manned space flights mean yet the greatest challenge, adventure and responsibility for everyone involved. Long duration flights on low Earth orbit space station, as well as manned Moon and Mars missions are ahead in the near or far future. Safety and health of the crew is one of the most important critical aspects of these missions. One of the major health concerns is the damaging effects of ionizing space radiation (Parker and West, 1973). Once the crew leaves the Earth's protective environment, they will be bombarded by radiations of varying energies and intensities. Adequate shielding will be required to protect the crew from this environment.

There is a series of special issues of the journal "Radiation Measurements" devoted to space radiation measurements (Benton *et al.*, 1990a; Benton, 1992a; Benton, 1994; Benton *et al.*, 1996a; and Benton, 1996b). Interested readers can find there reviews (for example Horneck, 1992) and the most advanced up to date results including the study of space radiation environment (radiation belts, solar and galactic cosmic rays), space radiobiology, and results from special flights such as the Long Duration Exposure Facility and the MIR space station. There is also an excellent collection of information relevant to space radiation and protection published in the report of Wilson *et al.*, (1991). The following short introductory discussion is based mostly on this later work.

Space Radiation Environment

Figure 1. Space radiation environment (Wilson *et al.*, 1991).
The types of particle radiations that occur in space are summarized in Fig. 1 (Wilson *et al.*,

1991). The most harmful components of these radiations are trapped electrons and protons in the Van Allen belts, solar flare protons, and galactic cosmic rays. There are both significant temporal and spatial variations. For example, trapped particles exist only in the geomagnetic field where mirror points lie well above the atmosphere; the solar wind can only be seen outside the Earth's magnetosphere; solar cosmic rays are rare transient events associated with solar flares; and so on. Electrons with energies below 100 keV and the protons below 10 MeV are mainly important only from a material point of view—for example, thermal control coatings—and are considered biologically unimportant. The radiations of immediate importance for biological consideration are the trapped protons in the inner zone, the trapped electrons in both the inner and the outer zones, and solar flare protons. Galactic cosmic rays are also biologically important. They are of low intensity but many questions surround them because of their particular composition, and their biological action is potentially hazardous and not well known experimentally.

Galactic Cosmic Rays

The galactic cosmic rays contain a little bit of everything. Fig. 2 show (a) the abundance and the associated tissue doses of galactic cosmic ray heavy ions as a function of atomic numbers (Simpson and Garcia-Munoz, 1991); and (b) the differential energy spectra of galactic cosmic ray ions for solar minimum conditions (Adams *et al.*, 1981).

(a)

(b)

Figure 2. (a) Galactic cosmic ray heavy ion intensities and associated tissue doses (Simpson and Garcia-Munoz, 1991). (b) Galactic cosmic ray ion spectra for solar minimum conditions (Adams *et al.*, 1981).

The dose is proportional to the charge squared. The relative dose contribution is more or less the same for different particle charges; it does not follow that the less abundant charges are necessarily negligible. Nevertheless, eight ions from hydrogen to iron contribute about 70% of the expected equivalent dose.

The galactic cosmic rays are affected by interaction with the Earth's magnetic field. Mainly the low rigidity (mass to charge ratio) particles are excluded from equatorial regions at low

altitudes, whereas near the poles the particles may come in freely at all altitudes. Although the particles with low rigidity are seen at low altitude mainly near the polar region, the heavy ions are by far the most rigid particles of the galactic beam. Consequently, mostly protons are lost in the equatorial region having a greater proportion of heavy ions.

The galactic cosmic rays coming in from galactic space interact with the interplanetary plasma, which is resulting from the expanding solar corona. The maximum intensity of the galactic cosmic ray flux occurs at solar minimum due to the lower solar plasma output.

Solar Cosmic Rays

Occasional solar flares are associated with solar activity. Plasma is ejected from a large number of optical flares, and during some of these events, there are particles that are ejected at high energy into interplanetary space. These high-energy particles are able to escape the solar magnetic fields only if the magnetic field lines are open to the interplanetary region. Generally there are few solar events which might be called “major events” during a solar cycle. Major events are usually absent during solar maximum and minimum.

(a)

b)

Figure 3. (a) Solar cosmic ray Earth interaction. (b) Solar cosmic ray proton fluence of three major solar events.

The solar cosmic rays produced on the Sun may travel to the Earth. The transit time between the Sun and the Earth is typically 20 minutes for relativistic particles, but sometimes it takes up to a few hours depending on the energy spectrum and the interplanetary magnetic field structure (see Figs. 3a and 3b).

The spectral distribution at Earth changes as a function of time because high-energy particles tend to arrive before lower energy particles. The angular distribution of the particles varies greatly from event to event. During some of the major events, the particles tend to be directional early in the event and approach isotropy later as the lower energy particles arrive.

Similar to galactic cosmic rays, the solar protons tend to be eliminated from equatorial regions of the Earth's magnetosphere. However, nearly all particles incident in polar regions are transmitted to low altitudes.

A reliable method of estimating the size (integrated intensity) of events and which provides adequate real-time protection does not yet exist. In addition, the biological effects of protons, which give the dominant source of risk from solar particle events, are not well understood.

Geomagnetically Trapped Radiations

The trapped radiations, illustrated in Fig. 4., follow a helical path along the magnetic field lines between the mirror points. The location of the mirror points along the field line depends on the pitch angle at the magnetic equator and the energy of the particle. The greater the energy or the higher the pitch angle, the deeper the mirror points lie in the magnetic field. If the particle energy and pitch angle are sufficiently large, the mirror point is so deep that the particle interacts with the atmosphere and is lost from the particle population.

Figure 4. Near-Earth trapped radiation and solar proton environment (Parker and West, 1973).

The Earth's radiation belts contain substantial fluxes of electrons and ions. Earlier it was thought that protons were dominant ion species with only minor contributions of helium and heavier ions. However, when relative comparison viewed in terms of total ion energy rather than energy per nucleon heavier ion species can be very important. In such comparisons it is found that helium ions can compete favorably with hydrogen in relative abundance, and at MeV energies oxygen and iron ions can be very significant. In contrast, comparison of ionic composition generally tends to favor protons in the upper keV and MeV energy ranges when energy per nucleon is considered (Spjeldvik, 1996).

For the inner zone it appears, at least for the protons, that the particle sources are primarily neutrons, which are produced in atmosphere by solar and galactic cosmic rays. The outer zones appear to be something like a pipe line with strong sources and strong sinks. The

particles flow rapidly through these regions, and on the average they maintain a fairly high population density although the residence time is short. (See Singley and Vette, 1972.)

2.1.2. Radiobiological Issues

Current methods for evaluating dose equivalents resulting from heavy ion exposure utilize biological effectiveness quality factors (Q) which are specified as functions of linear energy transfer (LET) of the projectile particles to the biological system being traversed (ICRP-26, 1977; ICRP-60, 1990). While risks from low LET radiation can be derived from effects observed in the Japanese atomic bomb survivors, the data required to assess risks from high-LET space radiation are rare. This is particularly important since the quality factor of high LET radiation can be as much as 30 times greater than of an equivalent dose of low LET radiation. Other studies have suggested abandoning the 'Q value-LET system' (Katz, 1986) and formulating more detailed models of cell destruction and transformation using radio-sensitivity parameters derived from biological experiments (Cucinotta *et al.*, 1991). The largest uncertainty in space radiation risk assessment arises from the scarcity of radiobiological data. Much of the research on this field can be done at heavy ion accelerators on the ground. However, the question of whether radiation effects are amplified in zero gravity can be answered only by space research.

2.1.3. Dosimetric Measurements

Present space flight dosimetry instrumentation includes dosimeters of both passive (thermoluminescent (TLD), and etched track type) and active (proportional counter) detectors distributed throughout the Shuttle volume, and they have been shown to be reliable and accurate for the Space Transportation System (STS) missions (Atwell, 1990; Reitz *et al.*, 1996). TLDs are used to measure total absorbed dose. Etched track detector measurements are regularly used to measure the distribution of absorbed dose as a function of linear energy transfer (Benton, 1990b). Time-resolved radiation dosimetry measurements inside the crew compartment have been made during recent Shuttle missions with portable battery-powered tissue equivalent proportional counters capable of measuring the absorbed dose and dose distribution as a function of LET and time, for all penetrating radiation in space (Badhwar *et al.*, 1992; Golightly *et al.*, 1994). An intercomparison of radiation measurement was done in a joint NASA Russian study of the radiation environment inside the Space Shuttle on STS-63 (Badhwar *et al.*, 1996.)

In general, STS dose rates are relatively low. For the 28.35° inclination orbits at altitudes between 250 and 350 km, the average dose rate is observed to be approximately 0.1 mGy/day. Steady dose rates in deep space, even with shields, are expected to be substantially higher (factor of 5 to 10), with intermittent (solar flare) dose rates higher still. Further advancement in dosimetric instrumentation and techniques will be required to monitor the astronaut deep space exposures, with emphasis on active, as opposed to passive, dosimeters.

2.2. Radon Radiation Protection

In this section I will shortly discuss some basic scientific aspects of the “radon problem”. For more interested readers there are probably around a hundred of books and thousands and thousands of scientific papers published on the subject. Here, I should like to mention only three books that I have found myself very inspiring in my work. First is the book “Radon and Its Decay Products in Indoor Air”; edited by William W. Nazaroff and Anthony V. Nero, Jr. (1988). This book is a comprehensive source book on the incidence, effects, and control of indoor radon. It provides an authoritative, wide-ranging, and technically rigorous account of the occurrence of indoor radon, the nature and behavior of its decay products, health effects, and techniques and strategies for minimizing exposure to radon. The second publication is the Publication-65 of the International Commission on Radiological Protection (ICRP) on “Protection Against Radon-222 at Home and at Work” (1993). This issue of the ICRP summarizes the extent of current knowledge about the health effects of inhaled radon and its progeny and makes recommendations for the control of this exposure in both dwellings and workplaces. It gives guidance to national advisory and regulatory agencies and to practitioners of radiological protection concerned with radon in dwellings and workplaces. This ICRP publication is also available in Hungarian language (translated by Zs. Kertész [1996]). The third book is the “Element of Risk: the Politics of Radon.” written by Cole L. A. (1993). This book shows different aspects of the American experience coping with the radon problem. It rises fundamental political and ethical questions that even scientists, working on the field, cannot avoid. Concerning radon, certainly, a dangerous gas, these are not trivial things. After all hundreds of thousand’s or may be even million’s life is at stake.

2.2.1. Fundamentals of radon

The various isotopes of radon (^{219}Rn , ^{220}Rn and ^{222}Rn) are present to a greater or lesser degree in all environmental media—air, water, and soil—and arise naturally from the radioactive decay of radium, whose isotopes are members of the decay series starting with uranium and thorium, primordial constituents of the Earth’s crust. A principal characteristic of radon that gives it more radiological significance than earlier members of the decay chains is the fact that it is a noble gas. As such, once is formed in radium-bearing material, a radon atom is relatively free to move. Once in the pore space, macroscopic transport of radon is possible, either by molecular diffusion or by flow of the fluid in the pore space. Radon can therefore reach air to which humans have access. Formed in the ^{238}U decay chain from decay of ^{226}Ra , ^{222}Rn is the radiologically most important radon isotope because it has the longest half-life, 3.8 days. This is long enough that much of the ^{222}Rn formed either in building materials or in the ground within approximately a meter of building understructures can reach the indoor environment. In some cases ^{220}Rn (half-life 56 s) may also be important. ^{219}Rn (half-life 4 s) concentration in air is negligible. Typical activity concentrations of ^{222}Rn are 2-20 Bqm^{-3} in outdoor air, 10-100 Bqm^{-3} in indoor air, 1-10 kBqm^{-3} in underground cavities such as caves and non-ventilated mines, and 5-50 kBqm^{-3} in soil gas. The spatial distribution of these radon concentrations usually follows a lognormal distribution. Significantly higher levels than these averages may occur at startling frequencies.

The second important characteristic of radon is that it decays to radionuclides that are chemically active and relatively short-lived. As indicated in Fig. 5, the four radionuclides following the decay of ^{222}Rn have half-lives of less than 30 min, so that—if collected in the lung on being inhaled—they are likely to decay to ^{210}Pb before removal by lung clearance mechanisms. The radiation released on decay of the short-lived decay products imparts the lung dose to which increased risk of lung cancer is attributed.

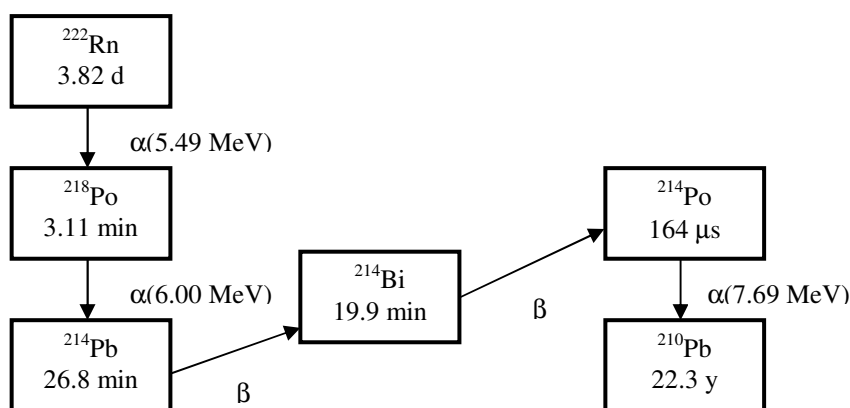


Figure 5. Part of ^{238}U decay chain containing ^{222}Rn and its short-lived decay products.

2.2.2. Sources and transport of indoor radon

Radon arises from trace concentrations of radium in the Earth's crust, and indoor concentrations depend on access of this radon into building interiors. Radon can enter directly from soil or rock that is still in the crust; via utilities such as water (and, in principle, natural gas) that carry radon; or from crustal materials that are incorporated into the building structure in the form of concrete, rock, and brick. The relative importance of these pathways depends on the circumstances, but it has become clear that the first—direct ingress from the soil—ordinarily dominates the higher indoor concentrations that have been observed in homes.

Although transport via molecular diffusion accounts well for observed fluxes from building materials and uncovered soils, diffusion alone cannot account for the total entry rates observed in single family houses. It appears that bulk flow of soil gas driven by small pressure differences between the lower part of the house interior and the outdoors can account for the efficiency with which radon from soil enters such homes. Pressure differences of only a few pascals arise from winds and indoor–outdoor temperature differences. Soil gas contains enough radon that, on the average, only 0.1% of infiltrating air would have to be drawn from the soil to account for indoor concentrations. Recently, advanced theoretical simulations of transport are helping to form a fundamental picture. These, together with observed pressure and soil parameters appear to be consistent with soil gas flow-rates and in some extent with measured radon concentrations.

2.2.3. Elements of risk and radon dosimetry

The concentration of short-lived decay products in air is given by a collective concentration. That is the potential alpha-energy concentration (PAEC), which is defined as the sum of the potential alpha-energies of each short-lived decay product atom present in a unit volume of air. The potential alpha-energy of a short-lived decay product atom is the sum of the energies of alpha particles that the atom emits until it decays into ^{210}Pb . PAEC is measured in Jm^{-3} . The equilibrium-equivalent decay product concentration (EEDC), corresponding to a non-equilibrium mixture of radon progeny in air, is the activity concentration of each decay product in radioactive equilibrium with each other that has the same potential alpha-energy concentration as the actual non-equilibrium mixture. The SI unit of the EEDC is Bqm^{-3} . The ratio of EEDC to radon concentration is the equilibrium factor (F). The basic measure of radon exposure is the potential alpha-energy exposure (PAEE), which is the product of the potential alpha-energy concentration and time. PAEE has SI units of Jm^{-3}s .

The principal basis for present concern about exposures to radon's decay products is the experience with lung cancer incidence among underground miners (ICRP-47, 1986). High fatality rates observed among miners as early as the sixteenth century (Haerting and Hesse, 1879) were only later ascribed to lung cancer (Ludewig and Lorensen, 1924). And it was only after recognition in the 1950s that excessive lung cancer rates were occurring among uranium miners that exposure history versus incidence indicated a relationship between added risk to lung cancer and exposure to ^{222}Rn decay products. The principal study groups have been underground uranium miners in the United States, Czechoslovakia, and Canada, and iron, zinc, and lead miners in Sweden. The results of these studies differ significantly among themselves. Taking an approximate median of these data the individual annual risk per unit radon exposure becomes $0.9 \times 10^{-6} \text{ y}^{-1}(\text{Jm}^{-3}\text{s})^{-1}$ (Nero, 1988). Assuming that this attributable risk applies to a 30 year period of expression, this implies an individual lifetime risk per unit exposure of about $2.8 \times 10^{-5} (\text{Jm}^{-3}\text{s})^{-1}$. (The ICRP-65 adopted a nominal probability coefficient [fatality] of $8.0 \times 10^{-5} [\text{mJm}^{-3}\text{h}]^{-1} = 2.2 \times 10^{-5} [\text{Jm}^{-3}\text{s}]^{-1}$). Uncertainty in this risk estimate—about a factor of 2 to 4—can have substantial implications when considering the importance of radon exposures and strategies for controlling them.

Numerous investigators have examined the applicability of dose-response data from the miner studies to environmental and, in particular, indoor exposures. In the last ten years or so, there have also been many studies aimed at detecting a correlation between the incidence of lung cancer and exposure to radon in dwellings. Some of these have shown positive correlation, but many have not. Samet (1989) and Stidley and Samet (1993) have made reviews of these studies. The typical result is the conclusion that the dose-response factor derived from the miner studies can be applied relatively directly to environmental exposures. Recently a case-control study from Sweden (Pershagen *et al.*, 1994) provided, although statistically weak, but some quantitative data to support this conclusion. As a result, a typical $180 \text{ Jm}^{-3}\text{s}$ lifetime radon exposure (associated with $54 \text{ Bqm}^{-3} \text{ }^{222}\text{Rn}$) of a member of the general public implies an associated lifetime risk of 0.5%. A figure speaks for itself.

These estimates are quite uncertain, because of uncertainty in both the primary studies of the miners and the applicability of those results to environmental exposures of the general population. One aspect of this applicability issue is that the average radon exposure just cited, $180 \text{ Jm}^{-3}\text{s}$, is well below the range where the epidemiological data have much statistical significance. On the other hand, this is not true of the higher levels found indoors. Lifetime exposure to $1000 \text{ Jm}^{-3}\text{s}$ (associated with $300 \text{ Bqm}^{-3} \text{ }^{222}\text{Rn}$) exceeds exposures typical of present-day underground uranium miners and is in an exposure range where the associated

lifetime risk, estimated to be 2–3%, assumes much more significance because there is no longer need to extrapolate to low doses.

Because most workers exposed to radon will also be exposed to other sources of radiation, the ICRP-65 suggests using a dose conversion factor from radon exposure to effective dose. This conversion has been obtained by direct comparison of the detriment associated with a unit effective dose and a unit radon exposure. The detriment per unit effective dose is 5.6×10^{-5} per mSv for workers and 7.3×10^{-5} per mSv for the general public (ICRP-60, 1990). The detriment per unit exposure to radon progeny is $8.0 \times 10^{-5} [\text{mJm}^{-3}\text{h}]^{-1}$ both for workers and the members of the public (ICRP-65, 1993). In terms of detriment, an exposure to radon progeny of $1 \text{ mJm}^{-3}\text{h}$ is equivalent to an effective dose of 1.43 mSv for workers and of 1.1 mSv for the members of the public. This difference is entirely due to the different detriment coefficients for effective dose in ICRP-60. The conversions obtained in this way are called conversion conventions. They are based on an equality of detriment, not on dosimetry.

2.2.4. Measurement and control techniques

Given the importance of exposure to ^{222}Rn decay products entails the problem of formulating a control strategy, which includes measurement and control techniques. Three techniques are widely used: (1) Real time monitoring for either ^{222}Rn or its decay products using highly portable self-contained instruments. Grab-sample instruments usually use a scintillation material to measure alpha particles emitted in a small collection cell (for ^{222}Rn) or from a filter (for the decay products). Ionization chamber and semiconductor based monitors may be used in passive diffusion or in active pumping mode. These measurements are only giving the concentration at a specific time. On the other hand, they can be used to study fast temporal variations of concentrations. (2) A charcoal sampler can be used to collect ^{222}Rn over a period of a day to a week, then sealed and returned to a laboratory, where the gamma radiation from decay products contained in the sampler can be measured (Cohen and Cohen, 1983). (3) An etched track detector can be placed in a place for periods from weeks to a year, then returned to the laboratory where the plastic detector material is etched to show tracks left by the passage of alpha particles from the decay of ^{222}Rn and its decay products (Alter and Fleischer, 1981). Such detectors have a filter permitting entry of ^{222}Rn , but not its decay products. This technique has the advantage of giving a long-term average concentration.

The techniques to control indoor ^{222}Rn correspond closely to the basic factors that affect concentrations. It is generally thought that source-strength reduction is the best control technique. Better barriers, sealants, and construction techniques can significantly reduce entry rates. Another approach reduces the air pressure immediately under the house, thereby offering an alternative route for the radon flux from the ground. However, there are also circumstances where increased ventilation rates are the most appropriate.

Finally, little can be done effectively without carefully framing various objectives and creating an overall strategy against the problem of radon at home and at workplaces. Improvements in scientific understanding of the behavior of radon, as well as of the health effects of exposures, are prerequisites for coping with this.

3. Materials and Instruments

In this chapter description of the materials and instruments that we have used in our experiments are given. Also the results of calibration and quality assurance measurements, those which I have done myself, are presented here.

For cosmic ray charged particle measurements on board of space satellites we have used etched track detectors of different types, mostly CR-39 and in some cases polycarbonate materials.

For long-term (weeks, months) measurements of average radon concentration we have also used etched track detectors, namely LR-115-II (cellulose nitrate) and CR-39. The LR-115-II was used in inverted diffusion cup geometry, while CR-39 was used in a small volume radon detector, called Radamon. The Radamon radon detector and its applications to measure radon in situ in air, in soil and in waters were developed in our group. For the measurement of fast (hourly, diurnal) variation of radon concentration in air, we used active electronic instruments based on ionization chamber (Atmos and AlphaGUARD PQ2000); silicon barrier semiconductor detectors (Dataquas); and in some cases scintillation detectors (Pylon).

Measurements of other parameters, related to the transport of radon in soils and in houses, were done with different types of sensors. These measurements included weather parameters and some soil properties. They will also be shortly discussed.

3.1. Etched Track Detectors

Etched track detectors, or in other name solid state nuclear track detectors, are such insulators (certain crystals, glasses and polymers), in which latent tracks produced by penetrating ions along their trajectories can be chemically etched into conical shape etched pits (Fleischer *et al.*, 1975). There are several books about etched track detectors and their applications in different fields. The one just cited is the book of Robert L. Fleischer, P. Buford Price and Robert M. Walker (1975) "Nuclear Tracks in Solids: Principles and Applications". Good introduction can be found in the book of S. A. Durrani and R. K. Bull (1987) "Solid State Nuclear Track Detection"; and also in the book of Spohr (1990) "Ion Tracks and Microtechnology: Principles and Applications". Perhaps the newest book in this field is the one to which my colleges: J. Hakl, I. Hunyadi and A. Várhegyi have also contributed. S. A. Durrani and R. Ilić (1997) edited this book and its title is "Radon Measurements by Etched Track Detectors: Applications in Radiation Protection, Earth Sciences and the Environment".

3.1.1. CR-39

In most of our experiments in the last decade we have used CR-39 type etched track detectors. CR-39 (Columbia Resin-1939 from Pittsburg Glass Co.) had been known for a long time before Cartwright B. G. *et al.* (1978) have introduced it as an etched track detector. CR-39's chemical name is polyallyl di-glycol carbonate (PADC); its element composition is $C_{12}H_{18}O_7$. It is a clear, transparent, rigid plastic of density 1.31 gcm^{-3} .

Among many known etched track detector materials CR-39 is by far the most sensitive material, which is capable of recording protons and alpha particles with a wide range of energies. Another advantage of it is the clarity of its surface after etching and the low background.

We have used different, commercially available, brands of CR-39. The MA-ND/ α was developed and produced in the Mechanical and Optical Corporation of the Hungarian Optical Works (Mátészalka, Hungary) in collaboration with the Institute of Nuclear Research (Debrecen, Hungary). It was produced between 1982 and 1990. TASTRAK is a product of Track Analysis Systems Ltd. (Bristol, United Kingdom). We have found these materials excellent for alpha particle detection and radon measurements. For space radiation measurements on the Long Duration Exposure Facility we used American Acrylic type pure and DOP doped CR-39 materials.

Etching of MA-ND/ α and TASTRAK was generally done in 6.25N NaOH solution at (70 ± 0.2) °C for 5 hours. Etching of American Acrylic CR-39 was done in 6.25N NaOH solution at (50 ± 0.1) °C for different etching times. Other etching conditions also occurred depending on the requirements by the actual experiments.

Calibration of CR-39

Alpha Particle Response

For alpha particle calibration purposes of CR-39, track detector pieces were irradiated by a thin ^{252}Cf fission fragment and alpha particle source. ^{252}Cf decays with alpha particle emission of energy of about 6.1 MeV. Track detectors were irradiated in normal air conditions either in near contact geometry with the source or by varying the distance between the source and the detector in order to obtain alpha particles with different energies. Generally about 10^5 alpha particle latent tracks per cm^2 were produced in the detectors.

6 to 9 MeV alpha particles were obtained from thin ^{212}Po sources, which I prepared by depositing daughters of ^{220}Rn on aluminum plates in a small volume (2 liter) thoron chamber. ^{220}Rn gas was obtained from ThO_2 powder.

In some calibration experiments we have also used the MGC cyclotron in Debrecen to obtain alpha particles up to 20 MeV. Aladár Valek designed a special irradiation chamber for this purpose. Accelerated alpha particles were back scattered from a thin gold foil, and then were let out from the vacuum chamber through a 22 μm thick stainless steel foil. I have used Al absorbers to reduce the energy of alpha particles.

Etched tracks were measured under optical microscopes. For track size measurements I have used high (40 or 100 times) magnification dry objectives, and a measuring ocular. 10–20 tracks were measured to define the average track diameter. The standard deviation was 0.1–0.2 μm .

To obtain the bulk etch rate (V_B) of the material, on each detector, exposed in near-contact to ^{252}Cf , minor axes (d_f) of 10–20, randomly selected, etched fission fragment tracks were

measured. Half of the average of these minor diameters was used to estimate the removed layer thickness: $h = \overline{d_f} / 2$. V_B was obtained as: $V_B = h / t_E$, where t_E is the etching time.

To obtain registration sensitivity ($S = V_T / V_B - 1$, where V_T is the track etch rate) of CR-39, diameter of 10 to 20 alpha particle tracks was measured on each sample. Circular looking tracks (indicating nearly normal particle incidence) were selected for measurement on the basis of a visual inspection. Sample standard deviation of these track diameters was used to find outsiders. The average of the remained track diameters was used for sensitivity calculation.

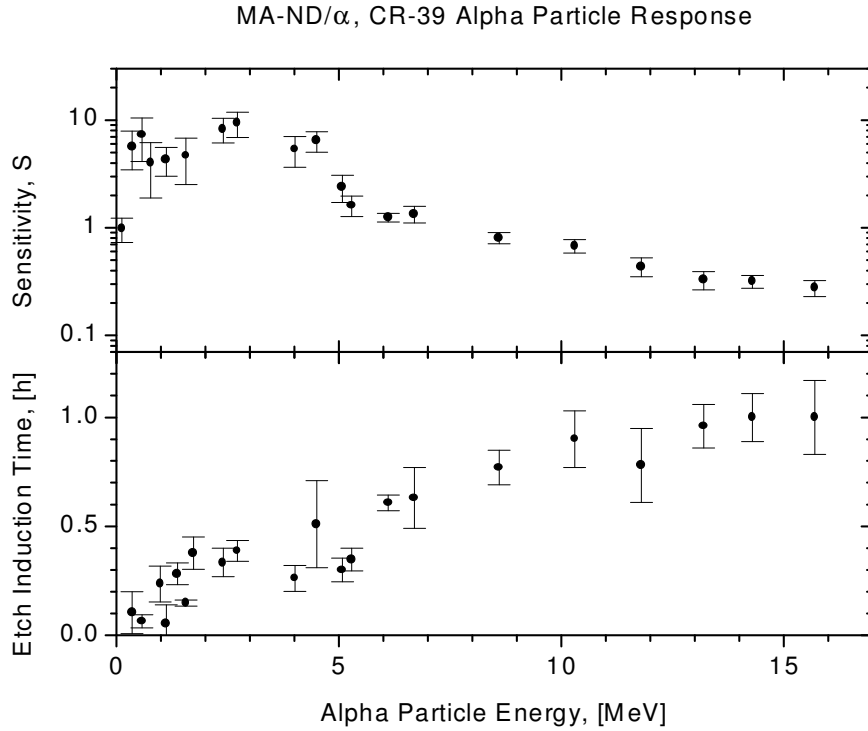


Figure 6. Alpha particle response of MA-ND/ α type CR-39 etched track detector.

In the case of the alpha particle response of CR-39, the concept (or model) of the so-called etch induction time was also considered. It means, the diameter of the etched alpha particle tracks (d_α) was measured as a function of etching time and then, the etching time was fitted as a linear function of the etched alpha particle track diameter: $T_E = a \cdot d_\alpha + b$, according to the least squares method (a and b are fitted parameters). Etch induction time (t_i) and the sensitivities were determined from the fitted parameters as: $t_i = b$ and $S = (1 + A^2) / (1 - A^2) - 1$, where $A = 1 / (2aV_B)$. Fig. 6 shows the alpha particle response of MA-ND/ α type CR-39 in the 1 to 16 MeV energy interval.

I have used these alpha particle response functions of CR-39 in detection efficiency calculations of CR-39 in different applications. This included the design of the Radamon radon detector (Csige, 1991a) and the application of CR-39 to neutron capture alpha particle autoradiography (Csige, 1993a).

LET Response of CR-39

The general response function of an etched track detector can be defined as the sensitivity of the detector as a function of a chosen response parameter. This response parameter is usually an energy loss type quantity, such as in the primary ionization, linear energy transfer or in the restricted energy loss (REL) models (Fleischer *et al.*, 1975). It is not known which quantity describes best the etchability of latent tracks. A model is generally chosen experimentally. The more the sensitivities for different ions lie on a single curve as a function of a chosen response parameter, the more the model is applicable for practical work. In our space radiation dosimetric measurement we chose linear energy transfer (LET) in water as a response parameter. Although it may not seem to make much sense why LET in water of different ions would be related to etch rate ratio in CR-39, the surprising fact is that it still works relatively well (see Fig. 7). The advantage of this practice was that CR-39 in this way could be directly used to measure “LET in water” spectra. LET in water is not very much different from LET in tissues as human body contains mostly water. LET in tissue could also be used but different kinds of tissues differ from each other almost as much as they differ from water.

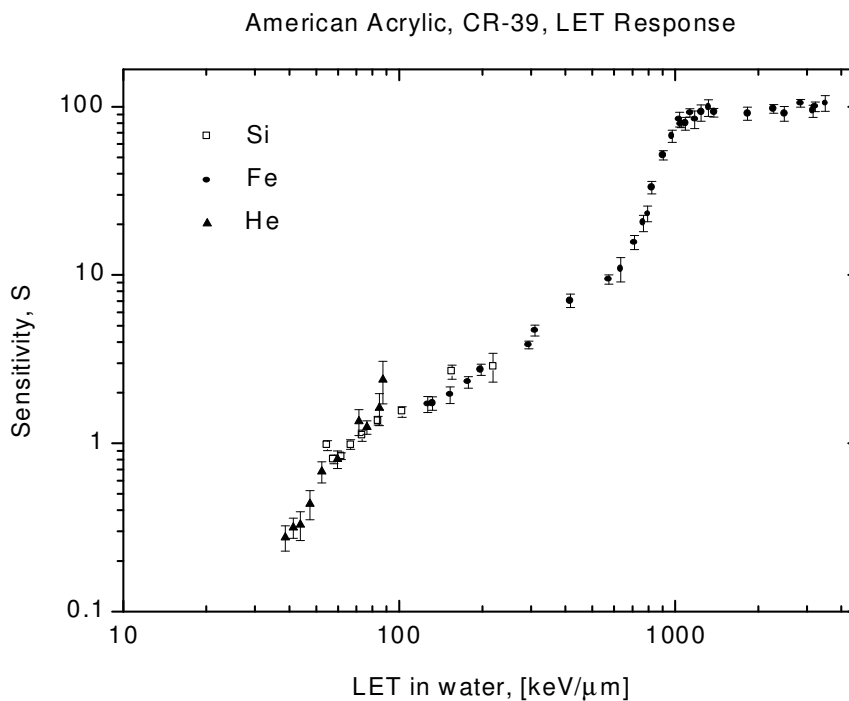


Figure 7. “LET in water” response function of CR-39 etched track detector. Sensitivity to different types of ions is plotted against linear energy transfer of particles in water.

Calibration experiments of CR-39 and polycarbonate etched track detectors with heavy ions were done at the Bevalac heavy ion accelerator in Berkeley (California, United States). The irradiations were done more or less regularly, though at random occasions. We usually had a chance to obtain a few minutes beam time when our collaborators had finished their work at around four in the morning. Stacks of etched track detectors were exposed to different heavy ions at different energies depending on what beam had actually been accelerated for our partners. 600 MeV Fe ions were easily available since it was used for therapeutic treatment in daytimes and the machine get used to it. Other ions were sometimes more difficult to obtain. To use the time more efficiently we used brass wedges in front of our detector stacks in order to obtain particles with lower energies. Stacks were irradiated at different incident angles, usually at 45°, 60°, 75°, and 90°.

Then the detectors were processed according to the standard used for the space irradiated samples. In some cases space and accelerator irradiated etched track detectors were etched together. For etched track size measurements we have used a videomicrometer. The microscope image of a track was captured by a video camera and track size measurements were made on the screen. Sensitivities (reduced etch rate ratio) less than about 5 were calculated from track opening diameters, while sensitivities greater than about 5 were calculated from etched track lengths. The model of constant etch rate ratio was used to calculate the sensitivities. No induction time was assumed.

Fig. 7. shows an example of the response function of CR-39 taken by different ions (He, Si, and Fe) with different energies. This figure shows that the sensitivity is a complex function of LET in water. It cuts down at low LET values and saturates at very high LET values. The removed layer was about 6 μm in this case. Lower LET particles (in the order of 10 keV/ μm in water) can also be easily detected with CR-39 at normal particle incidence and at longer etching time corresponding to about 40 μm removed layer. In our measurements of LDEF samples, however, only shorter etching times could be done because of the high track densities. Therefore higher portion of the LET spectrum could be studied.

Environmental Effects

Latent track formation in etched track detectors is the result of the prompt ionization around the particle trajectory and the subsequent physical-chemical processes in the damaged region. These later processes depend on the state of the detector material, which, however, can be affected by environmental conditions. As a consequence, sensitivity of the detectors may depend on the environmental conditions. On the other hand, the study of environmental effects may help to understand the nature of latent track formation, too (Benton, 1969).

I have studied the effect of vacuum, of CO_2 and of long term aging on the sensitivity of CR-39. In this point I will summarize the results of these studies.

Effect of vacuum

In the 1980-s, it was found in our laboratory, that the sensitivity of MA-ND/ α (CR-39) decreases dramatically, if the detector is irradiated in vacuum and is outgassed for a few hours prior to irradiation (Somogyi *et al.*, 1986). This effect must be seriously considered because track detectors are frequently calibrated by ion beams in the vacuum system of accelerators and used in air or vice versa. It may be especially important when CR-39 is exposed to space environment directly. I have investigated this vacuum effect in details, and we have published the results in a series of papers (Fujii *et al.*, 1987; Csige *et al.*, 1988 and Csige *et al.*, 1991b).

Samples of CR-39 detector of type MA-ND/ α were irradiated with alpha particles of different energies in normal air conditions and in vacuum using various pre- and post-irradiation storage times in vacuum. In the case of irradiation in normal air the distance between ^{252}Cf source and detector sheets was changed to obtain different alpha energies. For the same reason absorber layers of 6.3 μm thick mylar foil were placed between the source and the detector in the case of irradiation in vacuum. A magnetic shutter was used to control the irradiation in vacuum.

The main results and conclusions of these studies are:

1. I have found that the etch induction time increases and the sensitivity of CR-39 decreases significantly if the detector is irradiated in vacuum and is kept in vacuum for a few hours before and at least for a few minutes after the irradiation. No change in the bulk etch rate was found. I have associated the above characteristic times with outgassing of the oxygen from the sheets and with the stabilization of latent track in vacuum, respectively.
2. On the other hand, I have found that neither pre-, nor post-irradiation storage of CR-39 in vacuum changes the sensitivity of the detector, when the irradiation is done in normal air.
3. After a few hours of outgassing, the vacuum effect seemed to go to saturation. That is: CR-39 in vacuum has a significantly reduced (when compared to normal air conditions) but still stable response.
4. I have found no significant change in the sensitivity of CR-39, when storage time in vacuum before irradiation was not longer than a few minutes. This is important from the point of view of calibration when it has to be done in vacuum but the response is used in air experiments.
5. The ratio of sensitivity measured in vacuum to that measured in air seemed to be independent of the energy of the alpha particle in the 2-6 MeV energy region.

It was demonstrated by Drach et al. (1987) that the vacuum effect is actually the effect of lack of oxygen. In their experiment using a 2×10^4 Pa of oxygen environment the sensitivity to 840 MeV/amu U ions did not change in contrary to the case of 2×10^4 Pa of argon applied on the course of irradiation when the sensitivity decreased as dramatically as in vacuum. A possible explanation of these observations is that in the presence of oxygen free radicals produced by charged particles can combine with oxygen—resulting in a significant degradation of the material—, while in the absence of oxygen the recombination is more likely.

Effect of CO₂

SR-90 is a polymer produced from monomer ($m=1$) and oligomer ($m \geq 2$) of diethylene glycol allyl carbonate (Fujii *et al.*, 1988; 1993). Initial sensitivity of SR-90 just after polymerization is about 3 time higher than that of CR-39 (Fujii *et al.*, 1993). In the course of studies to develop this new polymeric track detector, M. Fujii *et al.*, (1995) have found that the amount of CO₂ dissolved in SR-90 plays a fundamental role in the latent track formation. It was also found that the sensitivity of CR-39 can be increased to some extent if it is aged in CO₂ of 1~3 atm at room temperature for several days.

From their experiments they have concluded that the sensitivity of SR-90 depends on the amount of CO₂ dissolved in the polymer at the time of etching. They have suggested that initial loss in sensitivity of CR-39 and SR-90 track detectors after their production may be due to outgassing of carbon dioxide (which was created inside the materials during polymerization) from the sheets.

On the other hand, at about the same time we have found, that irradiating CR-39 in pure carbon dioxide atmosphere drastically reduces the sensitivity of this material to alpha particles (Csige *et al.*, 1995b). I thought this effect was similar to vacuum effect (Fujii *et al.*, 1987) and was due to lack of dissolved oxygen in the detector material during irradiation, a gas seems to

be essential in latent track formation in CR-39. In a recent study (Csige, 1997a), however, I have confirmed that post-irradiation treatment of CR-39 in carbon dioxide increases its sensitivity. I have also studied the dependence of this effect on alpha particle energy, on the treatment time and on the pressure of carbon dioxide.

Experimental. I have used sheets of TASTRAK (Bristol, England), CR-39 type track detectors for the carbon dioxide sensitization experiments. Treatment in carbon dioxide atmosphere of CR-39 was made in a common household soda siphon at room temperature. The 2 liter volume metal vessel was dried, allowed to be filled up with fresh air, then CR-39 plastic detectors were introduced. Then the vessel was hermetically closed and carbon dioxide gas was introduced into it. The amount of carbon dioxide was estimated by measuring the mass of carbon dioxide patrons before and after usage. (Around 3% of the patrons released no carbon dioxide at all.) The vessel, in this way, became overpressurized. Partial pressure of air in it was always 1 bar, while partial pressure of carbon dioxide was around 1.5, 3 or 4.5 bars depending on how many patrons were used. The exposure time to carbon dioxide also varied from 1 hour to 1 week. After treatment the carbon dioxide was let out from the vessel. Then the vessel was opened and the track detectors were taken out. After treatment and before etching the detectors were kept in normal air for about 2 minutes. Etching was done in 6.25N NaOH solution at $(70 \pm 0.2)^\circ\text{C}$ for 5 hours.

The main results and conclusions of these studies are:

1. I have found that the sensitivity of CR-39 to 6 MeV alpha particles increased as a function of storage time in CO_2 and it was more than two times higher after a week than that of untreated sample. In other experiments, however, I have found that it was hard to reproduce this effect. One reason for this may be that I have used sheets of CR-39 from different shipments (first an old one, then a relatively new material). I have found that bulk etch rate does not change significantly when storing CR-39 in 3 bar carbon dioxide for a few days. However, after 1 week storage, I have observed a slight increase in bulk etch rate, too.
2. In another experiment I have studied the aging of CO_2 sensitized latent tracks. Treatment was done in 3 bar CO_2 for 1 week. Aging time varied between 2 min and 1 day. I have found slight decrease in the sensitivity of CR-39, which, however, was small comparing to the prior enhancement by the sensitization.
3. I have found that the sensitization effect of CR-39 by CO_2 does not depend on the age of latent track. The aging time varied between 2 min and half year, after which detectors were sensitized in 3 bar CO_2 for 160 hours.
4. In a yet another experiment I have studied the effect of pre-irradiation treatment of CR-39 in carbon dioxide. Samples were stored in 3 bar CO_2 for 160 hours. After this they were stored in normal air for 9 days. During this period the samples were irradiated with 6 MeV alpha particles at different times. The storage time between treatment and irradiation varied between 2 minutes and 9 days. I have found that sensitivity of pre-treated CR-39 was slightly higher than that of untreated samples, but the sensitivity increase was significantly smaller compared to post-irradiation treatment effect. This suggests that CO_2 has no significant effect on the bulk material to make it more sensitive. The sensitivity of pre-treated CR-39 did not depend on the storage time between the treatment and irradiation. This suggests that the amount of dissolved CO_2 in the polymer at the time of irradiation cannot be important in latent track formation, but it has an effect on existing latent tracks.

5. To study the LET dependence of the sensitization effect of CO₂ on CR-39 I have measured its sensitivity for alpha particles of different energies with and without treatment. Post-irradiation treatment was done in 3 bar carbon dioxide for 1 week. I have found that the ratio of sensitivity of treated CR-39 to untreated one does not show a clear dependence on the energy of alpha particle in the 2-9 MeV interval. Except at the lowest energy values (where the track diameter method is not very accurate in sensitivity calculation) this ratio was around 1.5 in this experiment. It suggests sensitization mechanism is not strongly LET dependent. This finding is very promising to extend applicability of CR-39 towards low LET cosmic ray charged particles and energetic neutron dosimetry applications.
6. In an experiment I have studied the effect of carbon dioxide sensitization of CR-39 on the calibration factor of our Radamon radon detector. (For description of Radamon see next section in this dissertation). I have found that calibration factor for radon measurement has not changed significantly with post-irradiation sensitization. However, etched track image quality improved significantly. Sensitivity of Radamon to thoron increased by about 20% when CR-39 was treated in carbon dioxide after exposure to thoron. The sensitivity of Radamon generally increases with CR-39 detector sensitivity. However, this increase saturates at certain etched track sensitivity. The geometry of Radamon was optimized for radon measurement using normal CR-39. Its sensitivity for radon cannot significantly be increased further by detector sensitivity increase. This, however, does not hold when the monitor is used for thoron measurement. Sensitivity saturation occurs at higher detector sensitivity for thoron than for radon.

From my experiments I have concluded, that latent tracks, formed in normal conditions, undergo irreversible transformation if they are exposed to carbon dioxide after irradiation. The transformed latent tracks are stable and have significantly higher track etch rate than normal tracks. My experiments, however, did not agree with the hypothesis, that dissolved carbon dioxide in the detector material during etching increases sensitivity (Csige, 1997a).

Effect of long term aging

In case of low activity measurements the irradiation time of etched track detectors can be very long, months or sometimes even years. The stability of the detector sensitivity during irradiation usually is expected, although it is not fulfilled in all circumstances. A favorable observation was published by Portwood *et al.* (1986) from this respect. They have found in a 4 months long experiment that the detector sensitivity during exposure does not depend on the time of particle incidence but only on the time elapsed between the production and the etching of the detector.

I have studied this question in 33 month long experiment and have confirmed the findings of Portwood *et al.* (1986) for this significantly longer period (Csige *et al.*, 1991b). This result means that the sensitivity history and the time of particle incidence has no importance during long time exposure, only a calibration is necessary at the time of the etching.

The sudden drop of sensitivity of CR-39 in a few weeks after manufacturing the sheets is well known. Less data were available, however, on the long term aging of this material. In a 3 years long experiment I have found that the sensitivity of MA-ND/ α (CR-39) decreased as a function of storage time before irradiation at room temperature (Csige *et al.*, 1991b). The

decrease was independent of alpha particle energy and was significant even after two years of storage.

I have also found that the sensitivity of, an initially 4 month old, detector stored at 4 °C did not drop during a 33 month long experiment, while that of stored at room temperature for the same time is reduced by a factor of 2 at all alpha particle energies. On the basis of this result it is recommended to store CR-39 detectors in refrigerator both before using and, if possible (for example in autoradiographic experiments), during long exposures.

In practice, if this is not possible to manage, a scaling factor may be introduced to characterize the relative sensitivity (and detection efficiency) of CR-39 to a standard one. This scaling factor has to be determined experimentally performing a calibration after any time of exposure when the detectors are etched.

3.1.2. Polycarbonate

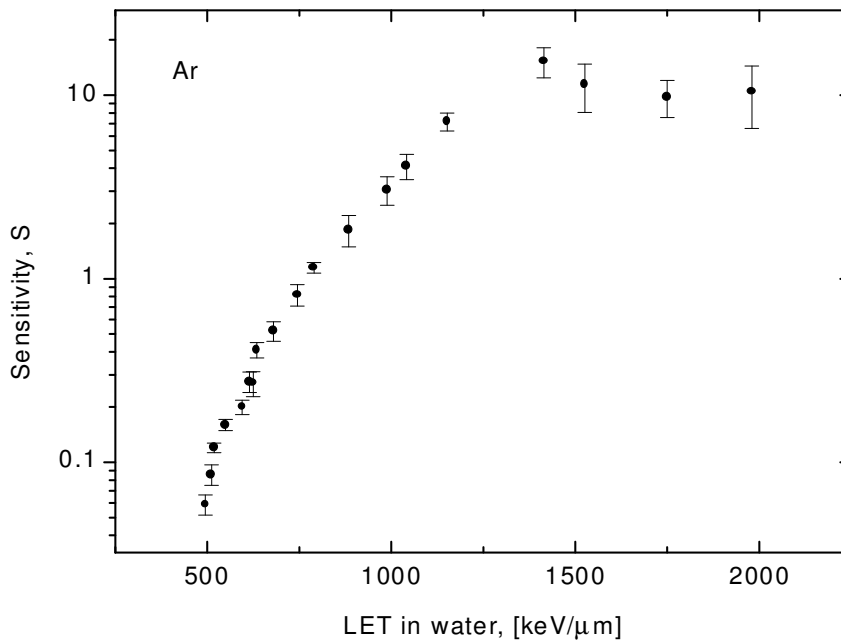


Figure 8. Response function of Sheffield type polycarbonate etched track detector. Sensitivity to Ar ions with different energies is plotted against linear energy transfer of particles in water.

In some cosmic ray charged particle measurements we have used two kinds of polycarbonate etched track detectors, Sheffield and Tuffak polycarbonates. These materials have much less sensitivity, than CR-39. They normally are not sensitive to alpha particles, only for heavier ions. This was an advantage, when we wanted to measure the high LET-tail of the LET spectrum of particles in track detector stacks exposed to cosmic ray charged particles in space. Fig. 8 shows the LET response of this material obtained by using Ar ions with different energies from the Bevalac heavy ion accelerator. In the LET range of 500 to 1000 keV/μm we preferred polycarbonate detector over CR-39 also because of its higher LET resolution in this range.

3.1.3. Cellulose Nitrate

In ^{222}Rn concentration measurements with the inverted diffusion cup technique, we used LR-115-II type (Kodak-Pathé, France) cellulose nitrate etched track detectors. This detector material has a nominal thickness of 13 μm, it is colored red, and it is layered on a 100 μm transparent support layer. This detector construction is used mostly for radon measurements. Cellulose nitrate is sensitive for alpha particles, and alpha particles between the energy region of about 1.5 MeV to 3.5 MeV can produce etched through tracks in the sensitive layer of the detector. Cellulose nitrate (LR-115-II) was etched in 2.5N NaOH solution at $(60 \pm 0.2)^\circ\text{C}$ for 150 minutes.

3.2. Radon Measurements

3.2.1. Diffusion Cup Detector

We have used inverted diffusion cup detector (Fig. 9) to measure radon concentration in air both in houses and in caves (Somogyi *et al.*, 1984).

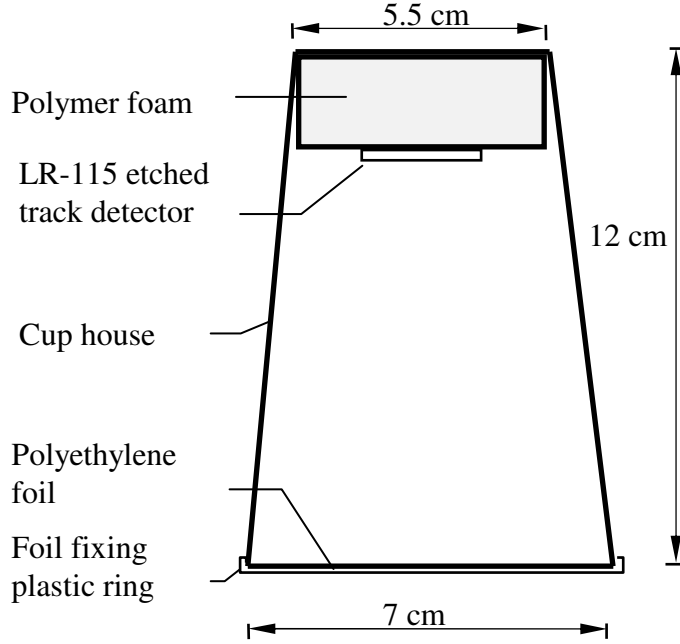


Figure 9. Scheme of the inverted cup used for radon concentration measurements.

The LR-115-II type (Kodak Pathe, France) etched track detector is fixed on a thermally insulating polymer foam disk by a double face adhesive tape. The cup opening is covered with a 35 μm thick polyethylene foil to keep aerosols, radon daughters and thoron (^{220}Rn) gas out of the measuring volume. This foil is fixed on the cup's opening perimeter with a plastic ring.

Radon diffuses into the measuring volume of the cup through the polyethylene foil. The foil causes a time delay in the temporal variation of inside radon concentration when compared to the external variation. Variation of internal ^{222}Rn concentration may be given as (Ward *et al.*, 1977)

$$\frac{dC_I(t)}{dt} = \frac{KA}{LV} [C_E(t) - C_I(t)] - \lambda C_I(t) , \quad (1)$$

where C_E is the external, and C_I is the internal ^{222}Rn concentration; L is the thickness of the foil; A is the area of the cup opening; V is the volume of the cup; and K is the radon permeability of the foil; λ is decay constant of ^{222}Rn ; and t is time. Putting the monitor into a field with C_0 external ^{222}Rn concentration with an initial condition of $C_I(t=0)=0$, the solution to equation (1) is $C_I(t) = C_0(\tau/\tau_M)[1 - \exp(-t/\tau)]$, where $\tau = (\tau_M\tau_R)/(\tau_M + \tau_R)$ may

be called as the transient delay constant of the foil (1.9 days); $\tau_M = (LV)/(KA)$ is the mean permeability time of the foil (2.95 days); and $\tau_R = 1/\lambda$ is the mean life-time of ^{222}Rn (5.52 days). Similar transient response is obtained when the external ^{222}Rn concentration suddenly decreases from C_0 to 0: $C_I(t) = C_0(\tau/\tau_M)\exp(-t/\tau)$.

It can be shown that if sufficiently long (more than a few days) exposure and post-exposure waiting times are applied, the transient effects at the beginning and at the end of the measurement period cancel each other. Otherwise a proper correction factor needs to be applied (Csige, 1991a).

In steady state conditions, radon concentration inside the cup is lower than outside. The cup geometry and the radon permeability of the polyethylene foil determine the inside to outside ratio of radon activity concentrations. I have found this ratio to be 0.65 for ^{222}Rn and 0.00024 for ^{220}Rn (Csige, 1991a). That means thoron is cut effectively from the measuring volume by the foil.

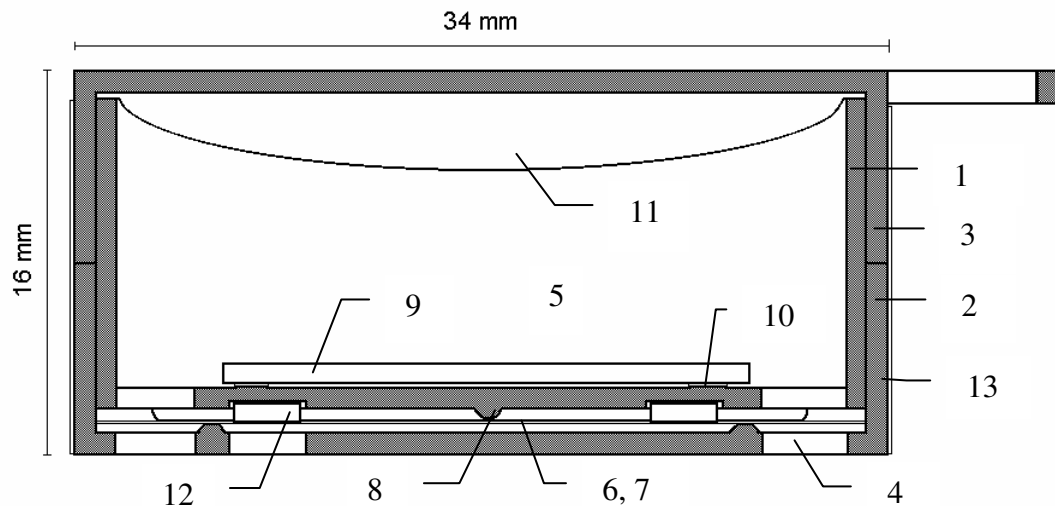
^{222}Rn atoms in the measuring volume decay and produce decay products. These in-cup born progeny tend to deposit on the walls of the measuring volume before they decay further. The direct product of ^{222}Rn decay is ^{218}Po which has a half-life of 3.11 minutes and a diffusion coefficient in air of $6 \cdot 10^{-6} \text{ m}^2 \text{ s}^{-1}$ (Bruno, 1983). With these numerical values I have solved the diffusion equation for ^{218}Po in the volume of this cup. I have assumed the volume activity concentration of ^{218}Po at the cup boundary surfaces to be vanished. I have found that in steady state conditions around 83% of ^{218}Po are deposited on the inner surfaces of the cup. The other short-lived decay products have even higher than this probability to deposit on the walls before decaying.

Alpha particles from the decay of radon and of its progeny can reach the detector surface to produce etchable tracks only from fractions of the volume and of the surfaces of the cup. These sensitive volumes and surfaces are determined by the energy of the alpha particles emitted by the different isotopes and the sensitivity of the etched track detector. The sensitivity of the monitor to radon exposure can be calculated accordingly (Somogyi *et al.*, 1984).

In practice, however, we used an experimental calibration factor. The calibration experiment was done in a radon chamber in Stockholm at the Swedish National Institute of Radiation Protection. It was arranged by I. Nikl from the “Frédéric Joliot-Curie” National Research Institute for Radiobiology and Radiohygiene, Budapest (Hungary) in 1988. Three times 5 cups were exposed to 10, 3, and 17 kBqm^{-3} ^{222}Rn activity concentrations for 8.8, 21.2, and 1.9 days, respectively. The detectors were evaluated using the standard procedure of our laboratory. I have determined the calibration factor to be $1 \text{ track}/(\text{mm}^2 \cdot 30 \text{ days}) = (60 \pm 7) \text{ Bqm}^{-3}$ of ^{222}Rn activity concentration (Csige, 1991a). Later intercalibration measurements have confirmed this experimental value. On the other hand, later experiments have also revealed some relatively large uncertainties due to non-perfect radon-tight closing of the polyethylene foil by the plastic ring. This uncertainty was reduced significantly in later measurements by using adhesive tapes instead of the rings to fix the foil more radon-tightly. Overall uncertainties of ^{222}Rn concentration measurements with this inverted diffusion cup detector, however, were found to be still relatively large, around 25% to 50%.

3.2.2. Radamon Radon Detector

The Radamon radon detector monitor was developed in the Institute of Nuclear Research (Debrecen, Hungary) in collaboration with the Mechanical and Optical Corporation of the Hungarian Optical Works (Mátészalka, Hungary) in the years of 1989-1990. Most of its development work I have done myself. The schematic view of the monitor is shown on Fig. 10.



- 1 Dosimeter house
- 2 Base
- 3 Cover
- 4 Quadratic arc shape openings
- 5 Sensitive volume
- 6 Paper filter
- 7 Polyethylene filter
- 8 Base and dosimeter house separator ribs
- 9 CR-39 etched track detector
- 10 Double face adhesive tape
- 11 Brass spherical calotte cupola
- 12 Thermoluminescent detector chips
- 13 Fixing tape

Figure 10. Schematic view and constituents of the Radamon radon monitor.

Radamon is a passive, diffusion type, integrating, etched track type radon detector. It is build up of three major plastic pieces. The cylindrical shape dosimeter house (1) is embedded between a base (2) and a cover (3). The base and the dosimeter house have four quadrant-arc shape openings (4), through which radon can enter the sensitive volume (5) of the detector. These openings have a width of 5 mm, and they can be aligned with the help of a pair of rib and groove, made on the dosimeter house and on the base, respectively.

The dosimeter house and the base are separated either by a 40 μm thick polyethylene foil (7) plus a paper filter (6), or by a paper filter (6) only. When both polyethylene and paper filters

are used, only ^{222}Rn can enter the sensitive volume of the detector with high probability. Ratio of internal to external radon concentration in steady state condition is 0.98 for ^{222}Rn and it is 0.02 for ^{220}Rn . On the other hand, when only paper filter is used, both ^{222}Rn and ^{220}Rn can enter the sensitive volume of the detector with high probability. Ratio of internal to external ^{220}Rn concentration in steady state condition in this later case is about 0.9. (I have obtained this later figure from identical ^{220}Rn exposures of three Radamons, in which 1, 2 and 3 paper filters were used, respectively. I have found the track density decreased as a function of the number of filter papers.)

Filters also keep aerosols and Rn progeny out of the sensitive volume. The paper filter is also good to lessen water condensation problems in humid environment. We use hydrophobic paper. In condensing environment water does not fill the pores of this filter easily; instead, water tend to form drops on the surface of it so, that the filter remains transparent for radon.

Fast response of the monitor depends on the surface (A), through which radon can diffuse into the sensitive volume of the detector. To enhance this surface, there are four pairs of ribs (8) on the inner surface of the base and on the outer (bottom) surface of the dosimeter house crossing each other pair-wise, in order to keep distance between the base and the dosimeter house. These ribs aimed to ensure radon diffuses through the filters on the whole bottom area of the dosimeter house. The surface to volume ratio is then: $A/V = 1.1\text{ cm}^{-1}$. Fast response of the monitor also depends on the radon permeability of the polyethylene filter, which was $K = 1.3 \cdot 10^{-9}\text{ cm}^2\text{ s}^{-1}$ in our case (Hakl *et al.* 1991). Corresponding delay time is $\tau = 2.7$ hours.

We used CR-39 type (MA-ND/ α or TASTRAK) etched track detectors in Radamon. The rectangular shape (usually $1.4 \times 1.4\text{ cm}^2$ or $1 \times 1\text{ cm}^2$ in size) track detector (9) is fixed on to the bottom of the dosimeter house using four small pieces of double face adhesive tapes (10). These tapes are applied at the corners of the detector. In this way, a second sensitive volume between the dosimeter house and the bottom surface of the CR-39 detector is formed. This means, both surfaces of CR-39 might be used for measurement. Normally, only the top surface is used, but in case of extremely high radon exposure, when track density on the top surface is too high, it may still be countable on the button surface. In an experiment with 15 Radamons I have found that the ratio of track densities on the top to the bottom surface of the CR-39 track detector is 15.6 ± 1.5 .

A spherical calotte cupola (11), made of 0.3 mm thick brass, is facing the detector at the top of the dosimeter house. This cupola aimed at resulting in a uniform etched track density all over the etched track detector surface area, which is used for counting. This eliminates one source of uncertainty that may be present in such a small volume radon detector. The radius of the sphere was optimized by Monte Carlo calculation and also verified it with experimental measurements. In two experiments I have found the track density is uniform within 4% (which was the track counting statistical error) over a 2 cm distance across the CR-39 detector surface.

In addition to etched track detector, there are rooms for two chips of thermoluminescent detector (TLD) in Radamon. One of these chips is shielded by the dosimeter house, the other one is shielded only by the polyethylene foil and the paper filter. Equipped with CR-39 and two TLD chips, Radamon becomes a complex radon plus gamma plus beta dosimeter.

Calibration of Radamon

The Radamon radon detector has been calibrated both theoretically and experimentally. In the theoretical calibration procedure, first, I have calculated the deposition of radon decay products on the inner surfaces of the monitor by solving numerically the diffusion equation in the volume of the monitor. Then I have used Monte Carlo simulation to create large number of alpha particles (in the order of 40 000) from the decays of radon and its progeny atoms in the volume and on the surfaces of the monitor. The trajectory of each alpha particle simulated was tracked and checked if it hits the detector or not. Position of impact and the energy and the moving direction of the alpha particle at the point of the impact were calculated. Then the “Etched Track Computer (ETC) Code” was used to simulate an etched track for each particle impacted in the detector. (The ETC code was also developed by me. It is based on the theory of etched track development [Somogyi, 1980].) Variable etch rate ratio model was assumed. The ETC code first investigated if a latent track is etchable or not and if it was etchable then characteristic sizes of the etched tracks were calculated. Then etched track selection criteria, appropriate for microscope scanning and track recognition experience of human eyes, were introduced and the detection efficiency was determined.

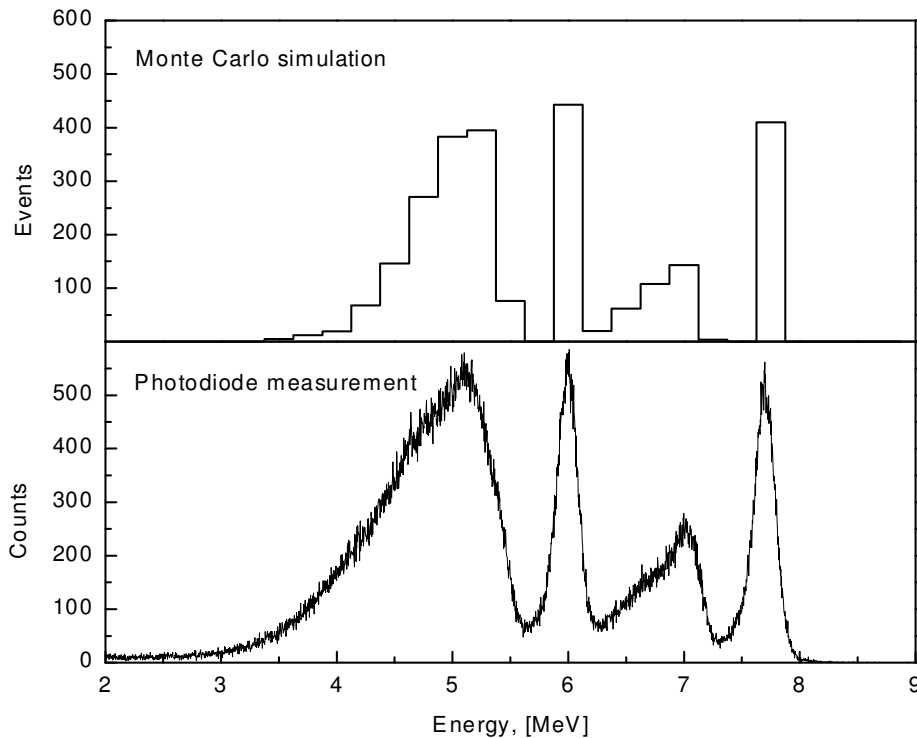


Figure 11. Monte Carlo simulated and measured energy spectra of alpha particles on the surface of the detector in the Radamon radon monitor. Measurement was done by J. Hakl substituting the CR-39 etched track detector with a Hamamatsu photodiode in a Radamon radon detector.

This procedure was used to design the monitor rather than for actual calibration. As a partial result of the calculation Fig. 11 shows for example a comparison of the Monte Carlo simulated and a measured energy spectra of alpha particles on the surface of the etched track detector in the Radamon radon monitor. Measurement was done by J. Hakl in 1992 substituting the CR-39 etched track detector with a Hamamatsu photodiode.

Experimental calibration of the Radamon was done in two different radon chambers. First, it was calibrated at the National Radiological Protection Board's (NRPB, Chilton, United Kingdom) radon chamber as part of the 1991 CEC international intercomparison of passive radon detectors (Whysall *et al.*, 1996) This calibration was arranged by my colleagues I. Hunyadi and J. Hakl.

Second, in 1993, I have calibrated Radamon in our radon barrel. Our radon barrel is a relatively small volume (0.14 m^3) cylindrical shape plastic barrel with height of about 80 cm. A ^{226}Ra source emanating ^{222}Rn can be placed approximately at the center of the barrel. After the barrel is closed ^{222}Rn concentration starts to build up in it. Different ^{222}Rn exposures can only be obtained by varying irradiation time. In this calibration experiment I measured ^{222}Rn exposures with Dataqua semiconductor based radon monitors, which were previously calibrated in a yet another radon chamber at the Swedish National Institute of Radiation Protection (see section 3.2.4. in this dissertation for the description and calibration of Dataqua instruments).

We have found that the experimental calibration value is significantly smaller than was predicted by the Monte Carlo simulation. There were also some discrepancies between calibration values obtained from the (1991) NRPB's and from my radon barrel experiments. In 1994 we have obtained an AlphaGUARD radon monitor. In intercomparison with its measurement I have confirmed the calibration value that I have got in my previous radon barrel experiments. Since that, we have been using this value as the standard calibration factor of the Radamon. It is $1 \text{ track}/(\text{mm}^2 \cdot 30 \text{ days}) = (84 \pm 6) \text{ Bqm}^{-3}$ of ^{222}Rn activity concentration.

In 1995 we have participated in two more international intercomparisons. One was arranged by Gilbert Jönsson from the University of Lund (Sweden) for the collaborating partners of an EU project. It was done at the radon chamber of the Swedish National Institute of Radiation Protection. The other one was the 1995 European Union intercomparison of passive radon monitors at the NRPB organized by Miles *et al.* (1996). Using the above experimental calibration value our measurements were within 7% of the nominal radon exposures reported by the organizers. I have used this 7% deviation as the relative error in our standard calibration factor.

Fig. 12 shows the results of a linearity test of Radamon. Five sets of Radamons with six in each set were exposed to different times in the barrel. ^{222}Rn exposures were measured with Dataqua instruments. Fig. 12 shows that the deviation of the signal of the Radamon from the predicted linear curve is very small, especially at higher ^{222}Rn exposures.

I have done a similar linearity test of the Radamon in a ^{220}Rn (thoron) chamber. In this case only a paper filter was used, so thoron could diffuse into the sensitive volume of the monitor. ThO_2 powder was used as thoron source. The volume of the thoron chamber was 2 liter. Irradiation time varied between 2 and 720 hours. As the half-life of thoron is 56 seconds, it takes only a few minutes for thoron to build up to its saturation value after the chamber is closed. Hence the thoron exposure and the etched track density was expected to be proportional to irradiation time. Fig. 13 shows measured etched track densities agree with this assumption within a factor of about 2. The straight line in Fig. 13 was obtained by linear fitting, in which fitted line was forced to go through origin.

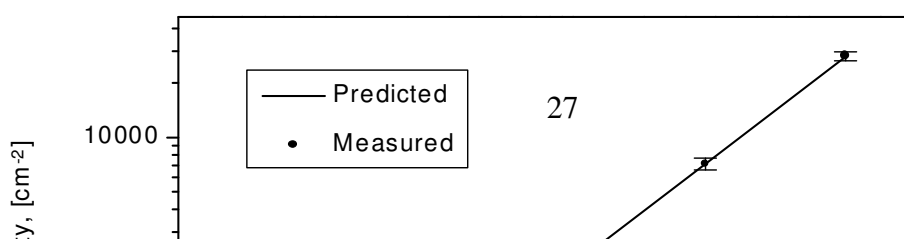


Figure 12. Comparison of predicted and measured etch track densities in a linearity test of Radamon. Irradiations were done in the radon barrel of the Institute of Nuclear Research. Prediction was made using the standard experimental calibration factor of Radamon. ^{222}Rn exposures were measured with a Dataqua semiconductor radon monitor.

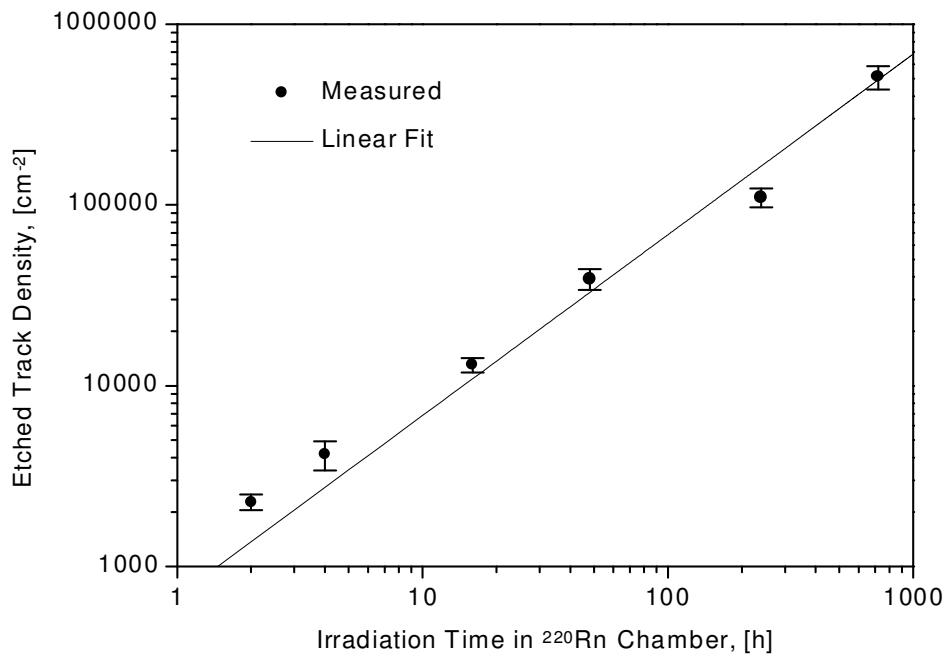


Figure 13. Linearity test of Radamon to ^{220}Rn (thoron). Thoron exposure is assumed to be proportional to irradiation time in the thoron chamber. The fitted line was forced to go through origin.

No signal was obtained in those Radamons, which had polyethylene filters in addition to the paper filters. This means two important things. First, that polyethylene filter effectively cuts ^{220}Rn out from the measuring volume. Second, that no detectable amount of ^{222}Rn were present in the thoron chamber during the irradiation. This result is an experimental

verification that a doublet of Radamons (one with paper filter only and one both with paper and polyethylene filter) can be used to measure both ^{222}Rn and ^{220}Rn concentrations.

I have estimated the sensitivity of Radamon to ^{220}Rn to be about 10-20% lower than to ^{222}Rn ; first because of the 10% reduction of thoron by the filter as I have discussed earlier, and second, because of the higher energies of alpha particles emitted by ^{220}Rn and its decay products. (CR-39 has less sensitivity to higher energy alpha particles.)

However, Radamon has not yet been experimentally calibrated to ^{220}Rn . In our measurements we have used the following estimated (or more or less guessed) calibration factor: 1 track/(mm² 30 days) = (100±30) Bqm⁻³ of ^{220}Rn activity concentration. (Standard error of the calibration number was estimated as the average deviation of the measured values on Fig. 13 from the linear curve.) Because Radamon with paper filter measures thoron as a supplement to ^{222}Rn , lower detection limit of measurable ^{220}Rn concentration depends on how much ^{222}Rn is present. In general we have found that detectable lowest concentration of ^{220}Rn is in the order of 25% of ^{222}Rn concentration.

3.2.3. AlphaGUARD PQ2000 Instrument

The AlphaGUARD PQ2000 is an ionization chamber type multiparameter ^{222}Rn monitor, frequently used in indoor environment to monitor ^{222}Rn and some other relevant parameters (most commonly atmospheric pressure, temperature and relative humidity) (Genrich, 1993). It has an active detector volume of 0.56 liter, which volume is separated from the environment by a glass fiber Rn-progeny filter. The sensitivity of the detector is 1 count per minute at 20 Bqm⁻³. The instrument's calibration error is 3 % (plus the uncertainty of the primary standard). It has a wide dynamic range of linear response between 2 - 2 000 000 Bqm⁻³. Linearity error is < 3 % within the total range.

The radon measurement integrating time is either 10 minutes or 60 minutes (user selectable). Data capacity is 20 days at 10 minutes integrating time and is 4 months at 60 minutes integrating time. It can be operated from internal battery for about 10 days.

The AlphaGUARD PQ2000 has temperature, pressure and relative humidity sensors inside its body box. Pressure is measured with a laser-trimmed silicon bridge-transducer sensor with resolution of 3 Pa in the 800-1050 hPa measurement range. Initial calibration uncertainty is ± 3 hPa. The temperature coefficient is typically 0.1 hPa/°C.

The ambient temperature is measured with precision monolithic integrated circuit with a resolution of 0.2 °C in the range of (-15) – (+ 60) °C. Initial calibration uncertainty is ± 1.5 °C. Relative air humidity (rH) is measured with a hydrophilic polymer film on hybrid type sensor with a resolution of 0.2 % rH in the 0 – 99 % rH range. Initial calibration uncertainty is ± 3 % rH. In addition the instrument has an external sensor channel, a relocation sensor and a mains power monitor.

We have used the AlphaGUARD PQ2000 mainly to monitor temporal variation of indoor radon concentration in two radon test-houses. This work aimed at studying dynamic behavior of radon in houses. In some cases we have also used it to measure radon exhalation rate from

soils. This later application required methodological development, which I will describe in the rest of this section. First, I have measured the response function of the radon monitor to a linear and to a step-function change of the external radon concentration using our ^{222}Rn barrel. Then I have developed a model to describe the transient response of the instrument. Finally I used this model for measurements of ^{222}Rn concentration in a vessel, designed to measure ^{222}Rn exhalation rate on soil surfaces.

Transient response of the AlphaGUARD

The active volume of the ionization chamber is separated from the environment by a glass fiber Rn-progeny filter. In a normal mode ^{222}Rn diffuses into the detector volume through this filter. It causes a time delay. ^{222}Rn concentration in the detector volume needs time to reach a new stable value after the monitor has been exposed to a new (different) level of external ^{222}Rn concentration. The transient response function to such a step-function increase in external ^{222}Rn concentration is described by Genrich (1993) as follows: > 30 % of a new level within 10 minutes; > 70 % within 20 minutes; and > 90 % within 30 minutes. These data are valid, when the new level is applied exactly at the beginning of the first cycle under consideration. I have used these data to calculate the k transfer coefficient, as it is described below.

I have used our radon barrel for this experiment. The instrument was placed at the bottom of the barrel. It was operated in normal diffusion mode (without using an external pump). Measuring cycle (measurement integrating time) was set to 10 minutes.

There were two successive experiments done. In the first one, the response of AlphaGUARD PQ2000 to an approximately linear increase of the external radon concentration was measured. This was done by keeping radon concentration in the barrel at low (about 10 Bqm^{-3}) level till the end of a 10 minutes measuring cycle. A few seconds before this cycle has finished the ^{226}Ra source had been introduced to the barrel. Then for 6 measurement cycles (1 hour) the barrel was kept closed. A few seconds after the 6th reading cycle has finished, the radon barrel was opened, and the instrument was taken out and put into low (about 10 Bqm^{-3}) radon concentration.

At this moment the second experiment started to measure the response of the AlphaGUARD PQ2000 to a sudden drop in the external radon concentration. 6 sampling periods (1 hour) were used in this experiment, after which the radon concentration displayed by the instrument decreased to about 100 Bqm^{-3} , still about 10 times higher than the background level. The results of the measurement are shown on Fig. 14.

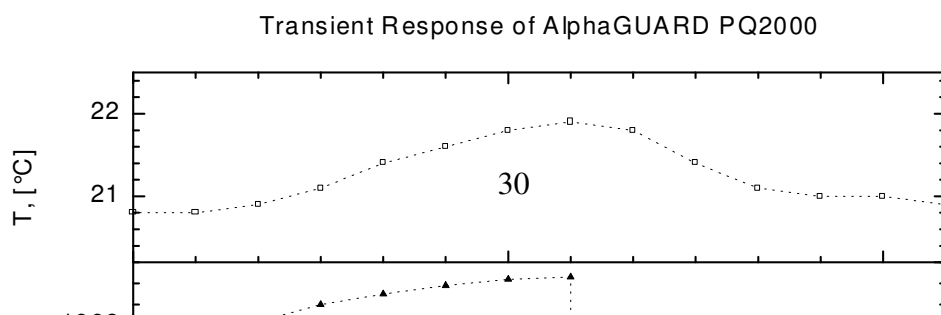


Figure 14. Response of AlphaGUARD PQ2000 to linear increase and then to sudden drop of ^{222}Rn concentration. Upper and middle parts of the graph show the temperature and the pressure variation as the instrument measured them. Dotted lines are shown only to guide the eye. On the bottom part of the figure the upper curve is the variation of ^{222}Rn activity concentration in the barrel, the lower curve is its variation in the ionization chamber. These curves are results of calculations obtained from fitting as described in the model of the measurement. Filled circles are the measured activity concentrations shown at the middles of the integrating cycles.

Fig. 14 shows that the pressure in the radon barrel became higher than outside the barrel at the moment the barrel was closed. After this both the temperature and the pressure increased inside the barrel. The pressure drop at the time of the opening of the barrel (a few seconds after 19:10) is higher than the increase at the closing. I have found that this increase in the pressure difference can entirely be associated with the temperature increase during the measurement. This indicates that the barrel was closed airtight.

Model

I have developed a compartment model to describe the variation of radon concentration in the measuring volume of the ionization chamber. This model consists of two compartments, one for the radon barrel, and one for the ionization chamber of the AlphaGUARD PQ2000. The variation of radon activity concentration in the ionization chamber (C_I) is assumed to be governed by the differential equation:

$$\frac{\partial C_I(t)}{\partial t} = k(C_E(t) - C_I(t)), \quad (1)$$

where C_E is the external radon activity concentration and k is a transfer coefficient. The decay of ^{222}Rn in the ionization chamber is neglected.

Let us consider first the case, when the external radon activity concentration suddenly increases from a low (negligible) value to a new constant value; that is $C_E(t < 0) = 0$, and

$C_E(t \geq 0) = C_0$. If $C_I(t = 0) = 0$ then the solution to equation (1) is $C_I(t) = C_0(1 - e^{-kt})$. If the first measuring cycle starts at $t = 0$, then the average radon activity concentration inside the ionization chamber in the i -th measuring cycle ($\overline{C}_{I_i}$) is given as:

$$\overline{C}_{I_i} = \frac{1}{T} \int_{(i-1)T}^{iT} C_I(t) dt = C_0 \left\{ 1 - \frac{1}{kT} e^{-ikT} (e^{kT} - 1) \right\}, \quad (2)$$

where T is the length of a measurement cycle. Using this equation and the transient response function data in the form, as it was given by Genrich (1993), the transfer coefficient could be estimated. Least squares best fit gave $k = 0.0014 \text{ s}^{-1}$.

Let us consider now the case, when $C_E(t) = \alpha t$, where α is a constant. With the initial condition of $C_I(t = 0) = 0$, the solution to (1) is $C_I(t) = \alpha \left[t + (e^{-kt} - 1) / k \right]$. The average radon activity concentration in the ionization chamber in the i -th measuring cycle can be calculated as $\overline{C}_{I_i} = \alpha \tau_i$, where

$$\tau_i = \frac{1}{k^2 T} e^{-ikT} (e^{kT} - 1) + \left(i - \frac{1}{2} \right) T - \frac{1}{k}. \quad (3)$$

These equations can be used to estimate the parameter α using the least squares method. If C_i is the measured average radon activity concentration in the i -th measuring cycle, and σ_i is its standard error, then minimizing the square sum of $S(\alpha) = \sum_i (\alpha \tau_i - C_i)^2 / \sigma_i^2$ results in

$$\alpha = \frac{\sum_i \tau_i C_i / \sigma_i^2}{\sum_i \tau_i^2 / \sigma_i^2}. \quad (4)$$

In the case when $C_I(t = 0) = C_0$ and $C_E(t > 0) = 0$ the solution to equation (1) becomes $C_I(t) = C_0 e^{-kt}$, and the average radon activity concentration in the ionization chamber in the j -th measurement cycle is $\overline{C}_{I_j} = \beta(k) e^{-k j T}$, where $\beta(k) = C_0 (kT)^{-1} (\exp(kT) - 1)$. If C_j is the measured average radon activity concentration in the j -th measurement cycle, then we may use these equations to fit the parameters C_0 and k .

Application to ^{222}Rn Exhalation Rate Measurement

^{222}Rn exhalation rate (or ^{222}Rn flux density) measurements on soil surfaces can have a great importance in characterizing the radon source potential of a given site. For ^{222}Rn exhalation rate measurements usually an airtight, sharp-edged cylindrical exhalation vessel is inserted at a few cm depth into the ground. ^{222}Rn , exhaled from the ground, enters the vessel and its concentration in it grows up with time. The initial (a few hours long) part of ^{222}Rn accumulation in the vessel is quasi-linear and the steepness of its growth is determined by the exhalation rate from the ground. At the beginning the process is not sensitive for the troubling processes such as “back-diffusion” and ^{222}Rn escape through the soil (Jonassen 1983; Samuelsson 1987). For that situation the value of ^{222}Rn exhalation rate (E_{Rn} , [$\text{Bqm}^{-2}\text{s}^{-1}$]) can

be calculated as $E_{Rn} = V / A \cdot \alpha = h\alpha$, where V (m³) is the volume, A (m²) is the opened area towards the ground, and h (m) is the height of the exhalation cylinder.

First test of the measurement method was some study on effects of meteorological parameters on ²²²Rn exhalation rate on the floor of a cellar and on undisturbed outdoor soil surfaces. The results of these measurements will be discussed in chapter 4.3.

3.2.4. Dataqua Radon Monitors

Dataqua radon monitors are diffusion chamber type electronic radon monitors, which use silicon based alpha particle detectors (PIPS or photodiode) to detect alpha particles emitted by ²²²Rn and its progeny in the sensitive volume (Várhegyi and Hakl, 1994). The Dataqua instruments have been developed by the Dataqua Electronic Ltd. (Balatonalmádi, Hungary) in collaboration with the Institute of Nuclear Research (Debrecen, Hungary) and the Mecsekurán Mining Ltd. (Pécs, Hungary). We have got the idea of the silicon detector based environmental radon monitor and the first photodiode sensors from the Laboratory of Hydrogeology of the University of Montpellier (Montpellier, France).

In our experiments, we have used two types of Dataqua radon monitors. The so-called single-channel instrument measures only radon concentration, while the multi-channel monitoring system can measure three additional parameters (for example temperature, pressure, water level or water conductivity) simultaneously with radon concentration. A field data-logger is built in into the instruments, which can store about 3500 readings per channel. With a typical one hour reading cycle this allows about four months automatic measurement period. As the energy consumption of the instruments is extremely low this long measuring period can actually be achieved. (The single-channel instruments can operate from 4 AA size alkaline batteries for around 1 year.)

The instruments have cylindrical chambers with internal diameter of 5 cm and height of 12 cm. The chamber is used in inverted cup geometry. The silicon detectors are placed at the top of the chambers. the single-channel radon monitor uses a Hamamatsu photodiode with sensitive area of 1 cm², while the multi-channel instrument uses a Canberra PIPS detector with sensitive area of 3 cm².

Cross-sections of the single- and multi-channel Dataqua radon monitors are shown in Fig. 15.

Radon diffuses into the cylinder passively from the opening of the chamber at the bottom. Those alpha particles are counted, which have energy between a preset low-threshold (about 1.0 MeV) and high-threshold (about 8 MeV). Sensitive volume (the region from which alpha particles can reach the detectors with such energies) is limited to the upper 4-5 cm of the cylinder. Detected alpha particles include both emitted by the decay of radon in the sensitive volume of the cylinder, and those emitted by the progeny of radon in the sensitive volume and on the surfaces of the cylinder. A metallic plate is applied at about 3.5 cm distance from the silicon detectors to enhance counting sensitivity and to prevent the detector surface against moisture precipitation.

Figure 15. Cross-sections of the single- and multi-channel Dataqua radon monitors (Várhegyi and Hakl, 1994).

We assume that it is mostly ^{222}Rn concentration, which is measured by the Dataqua radon monitors. This is first because ^{220}Rn (thoron) has a too short (55 s) half-time to diffuse into the sensitive volume of the chamber. Second, because most of ^{222}Rn (and ^{220}Rn) progeny that enter through the opening of the chamber are supposed to be plated out on the lower part of the wall of the chamber, long before they could reach the sensitive volume. Hence, only ^{222}Rn atoms and ^{222}Rn progeny that are born in the sensitive volume will be present in the sensitive volume.

Calibration of Dataquas

Currently our group has 11 single and 5 five multiparameter Dataqua radon monitors. Some of these instruments were calibrated in the radon chamber of the Swedish National Institute of Radiation Protection in 1992. This calibration experiment was arranged by Rolf Falk with the help of Leif Nyblon from the Environmental Laboratory of that institute. My colleagues J. Hakl and I. Hunyadi did the experiments. J. Hakl has determined the calibration factors for those instruments, which were used in these experiment. Later, in 1993 I have reevaluated these data and did some intercomparison measurements with other Dataqua instruments using

our radon barrel. I have found that the sensitivities of different instruments differ only slightly. The calibration factors I have obtained varied between 135 and 148 Bqm⁻³/cph for the single channel instruments and from 58.2 to 62.8 Bqm⁻³/cph for the multiparameter instruments (cph stands for counts per hour) with mean values of 140 Bqm⁻³/cph and 60 Bqm⁻³/cph, respectively. I have found the background of the Dataqua instruments were usually very low (in the order of 0.1 cph) but in some cases I have measured relatively high (2 cph for single channel instruments and 5 cph for the multiparameter instruments) backgrounds, too. The background of an instrument is expected to grow as a function of the integrated radon exposure to which the instrument is exposed as the long-lived decay product ²¹⁰Pb builds up on the bounding surfaces (including the detector surface) of the sensitive volume. Lower limit of detection (LLD) could be determined accordingly. With 50% relative accuracy LLD was in the order of a few hundred Bqm⁻³ using 1 hour integrating time.

Transient Response of Dataquas

Similarly to the AlphaGUARD PQ2000 instrument, I have measured the transient response of a multiparameter Dataqua instrument using our radon barrel. I have used 10 minutes integrating time. I have found that after a sudden drop of the external radon concentration the count rate of the instrument first suddenly decreased by about a factor of two in the first 10 minutes and after that it decreased exponentially with a half-time of about 35 minutes. The initial decrease was mainly due to the release of radon gas itself from the measuring volume, while the subsequent slower decrease of count rate was due to the decay of radon daughters on the bounding surfaces of the sensitive volume. Extrapolating this daughter count rate to the time when the external radon concentration was reduced to zero I could estimate that (64±8)% of the total count rate are supplied by radon daughters and accordingly 36% from radon itself in this radon monitor.

I have also developed a computer program to do deconvolution of the radon time series as measured with the instruments taking account for the slow decay of the daughters. However, I have found that at 1 hour integrating cycles (which was normally used in almost all of our measurements) this has not much effect on the measured time series. Therefore, I have not used this deconvolution procedure in the analyses of our Dataqua radon time series. It was good to know, however, that a peak obtained in one measuring cycle should be followed by a count not less than about one 8th of the count of the peak. This information was used in some cases to rule out artifacts, false signals that occasionally occurred. These signals were found to be caused mostly by electric-magnetic shocks such as lighting and switching on high voltage instruments.

3.3. Other Measurements

3.3.1. Weather Parameters

Although the AlphaGUARD PQ2000 and the multiparameter Dataqua instruments measured some meteorological parameters relevant to radon transport in soils, for a more general and comprehensive study we have used a Weather Monitor II type micro meteorological station (Davis Instruments Corp. Hayward, California, United States) at our radon test-house in

Erdőbénye (Hungary). This station is an automatic weather station, which can be used to measure practically all weather parameter we thought to be relevant for radon transport in soils and its entry into houses. Fig. 16 shows a typical installation of this monitor, which was very similar to our installation.

Figure 16. Typical installation of the Davis Weather Monitor-II.

3.3.2. Gas Yield and Soil Properties

From a standard single-channel Dataqua instrument we have made another instrument to measure gas-flow rate from 20 m deep bore hole. For this purpose we have used a mechanical, two-chamber type gas-flow meter (normally used in households to measure earth-gas consumption). We have fixed a magnet on the rotating axis of the instrument, and used a magnetic switch to generate electric impulses for the counter. Lowest measurable gas-flux was about $Q_{\min}=200$ l/h. Below this gas-flux the overpressure of the gas in the bore hole (several hundred Pa) was not high enough to keep the flow-meter's mechanics in motion. The flow

meter worked well from spring to autumn of 1995, but it failed during the freezy winter of 1995-96.

Radium content of soil samples was measured by Z. Dezső from the Isotope Institute of the Kossuth Lajos University (Debrecen, Hungary) using gamma spectrometry.

I have determined soil porosity and soil water content from density measurements of wet and dried soil samples. The original (in-place) volume of samples were measured at the time of sampling by measuring the depth change of the cylindrical hole from which the samples were taken. I have dried the samples in an oven at 105 °C for 24 hours. Average density of soil particles was assumed to be 2700 kgm^{-3} in the calculations. (This value is a relatively universal constant in the Earth's crust.)

4. Results and Discussion

4.1. Space Radiation Measurements

In this section I will present some results that we have obtained in the Physics Research Laboratory of the University of San Francisco (San Francisco, California, United States) in 1991 and in 1992, when I worked there as a visiting scientist. We have got most of these results from the so-called Long Duration Exposure Facility (LDEF) satellite. We have published these results in a series of conference publications (Csige *et al.*, 1991c, 1992a, 1992b, 1993b; Benton *et al.*, 1991, 1992b, 1995a, 1995b; Nefedov *et al.*, 1995) and regular papers (Benton *et al.*, 1996c, 1996d; Nefedov *et al.*, 1996; Oda *et al.*, 1992, 1993; Shinn *et al.*, 1994). From these papers I will present only some selected results, those to which my contribution was decisive.

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The LDEF Satellite

The Long Duration Exposure Facility (LDEF) was a passive, free-flying, cylindrical satellite that carried a total of 57 experiments from 9 countries (Parnell and Benton, 1996). The satellite was about 9 m long, 6 m in diameter and weighed about 9750 kg when fully configured. The LDEF mission began on 6 April 1984 with the launch of STS (Space Transportation System) 41C and the deployment of LDEF into a 28.5°, 480×474 km orbit. According to the original plan, after a year in orbit the satellite would ride a shuttle back to Earth. But shuttle schedule slipped, then came the *Challenger* tragedy, and LDEF was stranded in orbit. Then the Sun erupted in violent storms. These heated and expanded Earth's atmosphere, which reached out, slowing LDEF in orbit so it sank Earthward. By January 1990 the satellite was falling fast, by around a km a day. Within weeks it would incinerate in the atmosphere like a giant shooting star. Then came *Columbia's* rescue mission and LDEF was returned to Earth by STS-32 on 20 January 1990. In a mission lasting 2114 days (5.787 years) LDEF completed 32 322 orbits and traveled a total of 1 187 086 139 km-s (Parnell and Benton, 1996).

Space radiation measurements on LDEF were done in 14 experiments at various locations and shielding depths. They included study of explosive nucleosynthesis of cosmic rays (O'Sullivan *et al.*, 1996; Domingo *et al.*, 1996), radiobiology (Hammond *et al.*, 1996), dosimetry and validation of radiation environment and transport models (Armstrong *et al.*, 1996; Watts *et al.*, 1996). Lacking a propulsion system, LDEF relied on gravity to hold a fixed position perpendicular to Earth surface. Because one end was 9 meter closer than the other to Earth's center of gravity, a slightly higher gravitational pull kept that end pointing Earthward. Precise placement of the trays by weight kept the spacecraft from spinning. With this the attitude of gravity gradient stabilized orientation of LDEF, the effects of directional characteristics of meteoroids, space debris, atomic oxygen and space radiations could be studied. In our experiments we have measured the directional characteristics of the trapped

protons (Nefedov *et al.*, 1995, 1996) and of Fe ions (Csige *et al.*, 1992b; Nefedov *et al.*, 1996). Adams *et al.*, 1991 and Barz *et al.*, 1996 studied directionality of other heavy ions. Primary objective of the ionizing radiation measurements made on LDEF was to compare these measurements with model predictions of the space radiation environment.

We had four etched track detector experiments on LDEF. The etched track detectors were arrayed in stacks inside hermetically sealed canisters. The A0015 experiment included canisters on both Earth and west (trailing) sides of the satellite. P0006 and P0004 experiments were on the west side and M0004 was on the east (leading) side of the satellite. The distribution of stacks over the satellite allowed different directions for particle incidence to be sampled. Although I contributed to all of these experiments, in this dissertation I will present results only from the P0006 experiment.

The P0006 experiment contained a central main and four side stacks (Fig. 17).

Figure 17. Block scheme of the P0006 experiment on LDEF. Directional measurements of galactic cosmic ray Fe particles were made in the main and side stacks. Trapped proton fluence measurements were made in the main stack.

The central main stack consisted of 9 sub-stacks of etched track detectors, the planes of which were perpendicular to the east-west direction. The side stacks were attached to the four sides of the main stack with the normal vectors pointing out from the main stack into space with the approximate directions as follows: north-space (side stack A), north-east (side stack B), earth-south (side stack C) and south-space (side stack D). (The orientation of the side stacks is assumptive in the sense that experimental results obtained are in agreement with and only with this assumption. Unfortunately it was not possible to confirm the orientation of the side stacks from the data available about the disassembly of the LDEF satellite.) In the surrounding of the P0006 experiment there were 4 other canisters of the P0004 experiment representing a significant amount of shielding from those directions.

The contents of the sub-stacks involved sheets of American Acrylic and DOP doped (Berkeley) CR-39, Sheffield polycarbonate and polyester etched track detectors, repeated at different depths through the stacks. Al-equivalent shielding thickness ranged from 0.65 gcm^{-2} to $> 11 \text{ gcm}^{-2}$. (Shielding represents only the vertical self-shielding of the experiments to space and not the total shielding from LDEF and other experiments.)

4.1.1. LET Spectra Measurements

LET spectra measurements provide information on the distribution of absorbed dose as a function of linear energy transfer (LET) of particles defined for a reference material. This is particularly important when LET dependent biological effects are concerned. We have measured LET spectra in terms of fluence (or mission average flux) of particles. Both primary cosmic ray and secondary (those that were produced in the detector material) particles were included in the measured spectra just as they both contribute to absorbed dose in tissues. Etched track detectors are directional sensitive so both local and directional dependence of LET spectra on LDEF could be studied (Benton *et al.*, 1996c).

We have used American Acrylic and DOP doped (made in Berkeley as an experimental production) CR-39 and Sheffield polycarbonate etched track detectors to measure LET spectra. For LET spectra measurements we used a standard track coincidence method and also some new methods, which I have developed especially to obtain the high LET portion of LET spectra with high statistical accuracy.

CR-39 measurements. In the case of CR-39 and the standard track coincidence method two adjacent surfaces of a CR-39 pair (reassembled in their exposure geometry) were scanned to detect track pairs formed by particles traversing the original (pre-etched) interface. This event selection discriminated against surface background and introduced a minimum particle range for registration of about $16 \mu\text{m}$. The outer surfaces of the CR-39 doublets were also examined to detect particles traversing the doublet to produce four aligned tracks. The track fluences were then separated into short range (16 to $2000 \mu\text{m}$) and long range ($> 2000 \mu\text{m}$) particles. The long range particles were mainly primary galactic cosmic ray (GCR) charged particles with projectile secondaries of $Z \geq 2$, while short range particles were mainly stopping trapped protons and secondaries emitted by target nuclei.

Etch rate ratio was determined from the measured minor and major axes of the track opening using a constant etch rate ratio assumption. This approximation is very good if the residual range of the particle is significantly greater than the removed layer. In our case, however, the residual range of the majority of particles was on the order of the removed layer. This means the constant etch rate ratio model is no longer valid and some systematic error is introduced when using it. This error widely depends on the range and on the dip angle of the particles and in some cases I have estimated it to be in the order of 50%. This causes an uncertainty in the LET determination even greater than that, i.e. a factor of about 2. This problem was partially overtaken using the multiple-etches technique.

For a more accurate measurement of the high LET portion of the LET spectrum with CR-39 (with some contribution from K. Oda from the Kobe University of Mercantile Marine, Japan) I have developed the multiple etches technique (Benton *et al.*, 1992b; Oda *et al.*, 1992). In the

multiple etches technique the growth curves of the track opening diameters are measured as a function of the etching time. Then these curves are fitted with functions derived from model calculations assuming variable etch rate ratio model. From the fitted parameters of these functions etch rate ratio at the reference surface (which was the pre-etched detector surface in our case) could be obtained. We have used this technique not only to measure LET but also to identify stopping protons and generate an internal calibration curve for CR-39. (In an independent experiment but with this very same technique we have observed slight dip angle dependence of proton response of CR-39 [Oda *et al.*, 1993].)

The multiple etches technique can also be used to measure the LET spectra from interval to interval. After a short etching time only tracks of high LET particles can be seen. Tracks of low LET particles are not yet etched because of the lower etch rate ratio and a possible etch induction time. I have found that these high LET particles in CR-39 were mainly short-range heavy recoils, but included stopping alpha particles, too. After longer etching times, the tracks of short-range recoils become over-etched and they can no longer be recognized as pairs. At the same time, however, pairs of lower LET particles appear and become measurable. With this technique, a large portion of the high LET tail can be covered. Shortest range of particles included in the spectra was determined by the shortest etching time, which is limited by the resolution of the microscope used to measure the track dimensions. The shortest ranged tracks measured were 2–3 μm in length. We have used four steps of etching. After each step of etching we have scanned the same detector area, mapped the newly appeared tracks and measured the sizes of track pairs in coincidence. After the fourth etching we measured the sizes of nearly 1000 etched track pairs. (Most of the scanning, mapping and measuring work I have done myself with some help from K. Oda and L. A. Frigo.)

The LET of the particles was calculated from etch rate ratios using external calibration curves. These calibration curves were generated using tracks of different heavy ions obtained from the Bevalac heavy ion accelerator at the Lawrence Berkeley Laboratory (Berkeley, California, United States). Finally, the LET spectra were generated using an assumed isotropic directional distribution of particles in the solid angle, from which particles could be detected. Detection solid angle of an etched track detector is directed perpendicular to the detector surface and its measure depends on the LET of incoming particles. Lowest LET particles included in the LET spectrum limit the maximum solid angle (which was in the order of 1 sr in our case) that can be used to generate the LET spectrum. Any unisotropy of particles within this solid angle is averaged over. Directional dependence of LET spectra was measured with stacks oriented toward different directions on the satellite (Csige *et al.*, 1993b; Benton *et al.*, 1996c). Directionality of individual cosmic ray particles, however, was studied in independent experiments (see sections 4.1.2 and 4.1.3 in this dissertation).

Polycarbonate measurements. Sheffield polycarbonate (PC) is a much less sensitive plastic etched track detector than CR-39, but has some advantages in measuring the high LET portion of LET spectrum. I have found its “LET in water” threshold is about 500 keV/ μm (see Fig. 14 in this dissertation), which means it cannot detect protons or alpha particles. Hence, the track densities in LDEF PC samples are much lower than in CR-39 samples and the overlapping of tracks is negligible. In the higher LET portion, PC has higher LET resolution than CR-39.

In a preliminary study, PC pairs were etched in 6.25 N NaOH at 50 °C for 40 hours to remove about 10 μm from each surface. Over-etched, bubble-like tracks were found on both adjacent surfaces, which I suspected to be tracks from short-range heavy recoil particles produced in collisions of high energy trapped protons with target nuclei. Later, I have confirmed this

assumption by comparing tracks in polycarbonate detectors that were exposed to high energy protons with energy similar to those in LDEF samples (Csige *et al.*, 1992a). The identification of these tracks in polycarbonate initiated the development of the special techniques to measure the contribution of these short-range heavy recoil particles to the LET spectra. In the case of polycarbonate, to be able to use the track coincidence method I have used a short etching time (12 hours), which removed about 3-4 μm from the surfaces. The range of the short range particles which can be identified as pairs using the track coincidence method is on the order of twice the thickness of the removed layer thickness and was about 6-8 μm in this case.

Again, LET spectra were generated assuming isotropic directional distribution of particles in the detection solid angle. This assumption, however, was highly questionable in this case because of the known strong anisotropy of the high energy trapped proton environment by which these recoils were produced. In accelerator calibration experiments with 200 MeV protons, I have found that the directional distribution of heavy recoils in polycarbonate reflects the directionality of the primary beam (Csige *et al.*, 1992a).

The high LET tail measured in CR-39 is considered to be more accurate than that in polycarbonate due to the shorter range particles included in the spectra, the more accurate determination of LET from measured track parameters in the CR-39 and to the inherent uncertainties in the polycarbonate measurements for the calibration and determination of LET.

Comparison of calculated and measured LET spectra

One of the principal objectives of measuring LET spectra on LDEF was to produce a set of spectra for comparison with calculated predictions using environmental models. During the mission a set of pre-recovery estimates of LET spectra for LDEF were calculated as a function of spacecraft shielding (Benton *et al.*, 1992c). Fig. 18a shows the predicted LET flux spectra under three shielding depths along with our flux measurements made in the P0006 experiment under similar shielding. The calculated spectra in Fig. 18a dramatically underestimated the LET spectra as compared to our measured results at high LETs. It is seen that the high LET tail is substantial and that the secondaries contribute an important fraction of dose equivalent.

The pre-recovery calculations were made by Derrickson (1992) using the Naval Research Laboratory, Cosmic Ray Effects on Microelectronics (NRL CREME) code of Adams (1986). A number of assumptions were made to simplify the pre-recovery calculations including the use of a spherical Al shielding model of LDEF. The altitude used was the initial LDEF deployment altitude. Secondary contributions to the LET spectra were neglected including 'projectile fragments' (secondaries from breakup of incident ions during nuclear collisions) and 'target fragments' (residual nuclei and secondary particles from stationary nuclei after nuclear collisions).

Inclusion of short-range high LET secondaries in the calculation of model LET spectra has been hampered by the complexity of the problem. The entire trapped proton energy spectrum needs to be propagated through the stopping material of the spacecraft and all elastic and inelastic interactions need to be estimated. On the other hand, measurement of short-range high LET secondaries has previously also been hampered by the difficulties of detecting these particles. With the development of the multiple-etches technique I was the first to be able to

measure the contribution of these particles to the LET spectra. The discovery of their relative importance then initiated efforts to include these particles, too in model calculations.

Preliminary post-recovery calculations of the secondary particle high LET tail by Armstrong (1995) (see in Benton *et al.*, 1996d) are plotted on Fig. 18b along with our measurements. Fig. 18b includes calculated LET spectra caused by nuclear recoils in three different minimum range categories—0, 8 and 16 μm —calculated for the P0006 stack geometry under 6.5 gcm^{-2} shielding. While the 16 μm curve is seen to fall well short of the measured coincidence spectrum above 100 $\text{keV}/\mu\text{m}$, it is substantially higher than previous calculations in the high LET region (Benton *et al.*, 1992c). Better agreement exists between the calculations for nuclear recoils with range $> 0 \text{ }\mu\text{m}$ and the measured LET spectra under 7.5 gcm^{-2} in polycarbonate and the 0.65 gcm^{-2} multiple etches spectrum (ranges $> 2 \text{ }\mu\text{m}$).

(a)

(b)

Figure 18. (a) Comparison of calculated pre-recovery LET spectra using NRL CREME code, based on spherical shielding model of LDEF at three shielding depths (Benton *et al.*, 1992c) with measured LET spectra from P0006 stack on the west side of LDEF (Benton *et al.*, 1996c). (b) Comparison of calculated post-recovery LET spectra from nuclear recoils with ranges greater than 0, 8 and 16 μm under 6.5 gcm^{-2} in the P0006 experimental geometry (Armstrong, 1995; Benton *et al.*, 1996d) with measured LET spectra from P0006 stack on the west side of LDEF (Benton *et al.*, 1996c).

The identification of short-range high-LET secondary particles, produced in interactions between trapped protons and the nuclei of the experimental detectors, as a major contributor to the LET spectra for LDEF type orbits is one of the most significant results gained from LDEF ionizing radiation experiments.

4.1.2. Trapped Proton Directionality and Fluence

Trapped protons encountered in the South Atlantic Anomaly contribute dominantly to the effective dose received by astronauts in low earth orbits. Therefore validation of trapped proton radiation environment models is of particular important. In this section I will present

the results that we have obtained in measuring mission fluence and directional distribution of protons in the P0006 experiment of LDEF. The mission fluence will be compared with model calculation.

I have developed the method to measure the directional distribution and the LDEF mission fluence of trapped protons with CR-39 etched track detectors in the P0006 experiment. Trapped protons entering at the top of the main stack of the P0006 experiment will stop at different places in accordance with their moving directions and energies. I have specified etched track selection criteria that limited selection to tracks of protons, which stopped in a well-defined detection layer thickness (h_{det}). Measurement of the stopping proton density and directionality at certain place makes it possible to obtain the differential energy fluence of protons from a certain direction and energy (E to $E + \Delta E$) interval. E is defined by the material thickness above the detector sheet, and ΔE is defined by the thickness of the detection layer in which the density of stopping protons is counted.

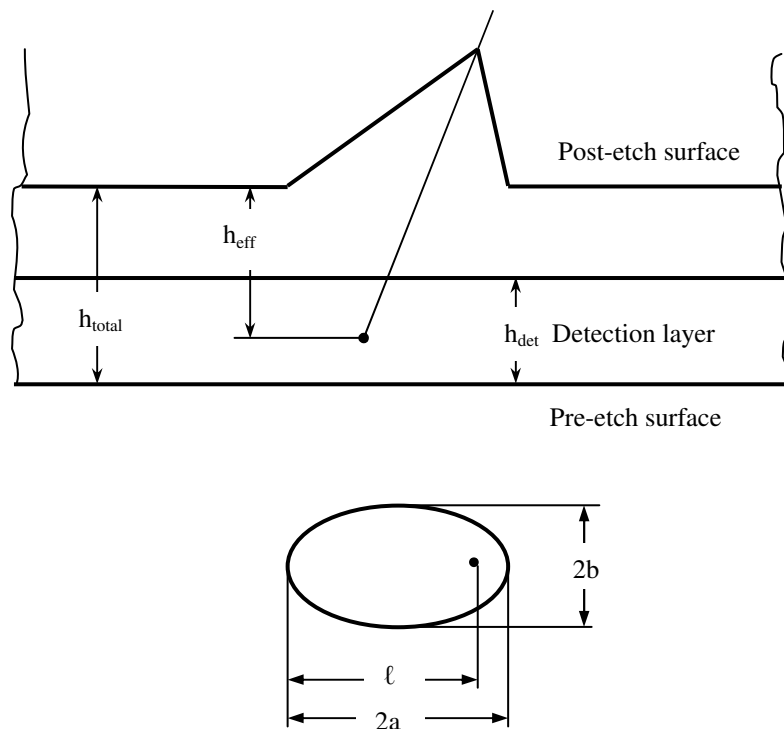


Figure 19. The measurable parameters of an etched track formed by a proton, which stopped in the detection layer near the bottom surface of the detector.

CR-39 sheets under shielding depths of 0.6 and 9.5 gcm^{-2} were chosen for these measurements. The detectors were processed for 36 hours in 6.25N NaOH solution at 50 °C for a total removed layer thickness of about 9 μm . After processing, two adjacent sheets of CR-39 were reassembled into their flight configuration on the microscope stage. Measurements were made at the centers of the sheets using the double sheet track anti-coincidence method. A particle event was selected for measurement when a pointed etched track was produced on the bottom surface of the top sheet but no corresponding track was found on the top surface of the bottom sheet. These selection criteria assured that a selected particle did not cross the original pre-etched surfaces of the detector sheets and that the total range of the particle was larger than the etched track length. Further selection of tracks was

made on the basis of the sizes of the tracks. The major and minor axes ($2a, 2b$) of the track opening and the distance between the “back” of the track opening and its tip (ℓ) were measured using a videomicrometer (Fig. 19). Only tracks with minor axes greater than a threshold were selected for evaluation. This selection criteria assured that the total range of selected particles were relatively large when compared to ranges of recoil particles. Tracks of most heavy recoil particles were over-etched and had rounded tips at their birth or stopping (depending on which direction the recoil is moved) point. Contribution of higher energy recoils and heavier than proton primary elements were negligible. Therefore it was assumed that stopping protons produced all tracks chosen by the above procedure.

A proton track of the selected type can be described by two parameters: (i) dip angle δ , the angle between the particle trajectory and the detector surface; and (ii) the effective removed layer h_{eff} , the distance of the particle stopping point from the post-etch detector surface (Fig. 19). To obtain these parameters from the etch track size measurements the theory of track development kinetics (Somogyi, 1980) was used. First, a constant etch rate ratio approximation (Henke and Benton, 1971) was tried but provided spurious results. This was not surprising since, near a stopping point of a particle, LET and the directly related track etch rate along the particle trajectory change rapidly. A variable etch rate ratio model of the form:

$$S = V - 1 = A \cdot REL_{200}^B, \quad (5)$$

was tried, where REL_{200} is the restricted energy loss rate in CR-39 with cut off energy parameter of 200 eV, A and B are parameters. Using equation (5) and my etched track computer code (based on the theory of etched track development) the values of a , b and ℓ were calculated as a function of δ and h_{eff} . As these functions were obtained only numerically, δ and h_{eff} were determined for each particle from a comparison of calculated and measured track sizes.

Its track to be selected a proton had to stop in the detection layer thickness h_{det} of the detector (Fig. 19). h_{det} is a function of the track size selection criteria. Under the given processing conditions only tracks with sizes greater than $4 \mu\text{m}$ were selected. From the detector response function [equation (5)] and the theory of track development this value defined the upper boundary surface (on Fig. 19) of the detection layer. Particles that stopped above this surface had tracks that were too small to be detected.

Since the detection layer was very thin and the scanned area was small, the volume density of stopping protons should be uniform. This meant that the distribution of the experimental effective removed layers should also be uniform. This criterion was employed to find the best values of the A and B parameters in the (5) detector response curve. Fig. 20 shows three examples of distributions of effective removed layers using different sets of A and B values. Since the maximum effective removed layer should be around $9 \mu\text{m}$ (which was the total removed layer), curve No. 2 was chosen in Fig. 20 and the thickness of the detection layer was determined to be $4 \mu\text{m}$.

Effective Removed Layer (h_{eff}), [μm]

Figure 20. Distribution of calculated effective removed layers using different detector response functions: (1) $V = 1 + REL^{1.4} / 162$, (2) $V = 1 + REL / 100$, (3) $V = 1 + REL^{1.1} / 432$.

The polar angle θ is defined as the angle between the particle trajectory and the normal of the detector surface in the main stack of the P0006 experiment. Tracks with polar angles $\theta \leq 30^\circ$ ($\delta \geq 60^\circ$) were selected for further analysis. This choice was determined to minimize the effect of scanning inefficiency at higher polar angles. Altogether 269 and 300 tracks were selected in sheets at shielding depths of 0.6 and 9.5 gcm^{-2} , respectively, in the main stack.

The arriving directions of the stopping protons for the CR-39 sheets at 0.6 and 9.5 gcm^{-2} shielding are shown in Fig. 21. It can be seen that the preferred direction of the arriving protons is not normal to the main stack. Both distributions have a maximum at polar angles of about 20° and at an azimuth angle that corresponds to the west direction. The distribution 0.6 gcm^{-2} is peaked more sharply, indicating perhaps some straggling of the protons as they passed through greater shielding.

To assess the eastward directed trapped proton fluences the tracks in the highest density quadrants near the west direction in Fig. 21 were chosen. There were 185 and 139 tracks found in these quadrants at the two increasing depths. Primary protons caused not all of these tracks. The contribution of secondary protons may be significant, especially at the greater shielding depth. In contrast, not all primary protons reach their stopping point without nuclear interactions. We have estimated the survival probability of a proton to be 97% and 86% for the two shielding depths. Assuming that all tracks are from primary protons and considering the survival probability, we estimated an upper limit of the average mission fluence of trapped proton fluence in the selected solid angle intervals. The corresponding energy intervals vary between 24 and 26 MeV for 0.6 gcm^{-2} and between 109 and 118 MeV for 9.5 gcm^{-2} shielding depths as the polar angles vary from 0° to 30° . The estimated average fluences are 6.3×10^6 and 9.2×10^6 protons/($\text{cm}^2 \text{MeVsr}$) at the 25 and 114 MeV mean energies. The relative error caused by counting statistics of these estimates is about 8%. Significantly larger error (about 25%) may be introduced for the estimation of the thickness of the detection layer. Other sources of error are most likely negligible compared with these estimates. The relative total error of the experiment is estimated to be about 30%.

0.6 gcm⁻²

9.5 gcm⁻²

Figure 21. Distribution of arriving directions of stopping protons at 0.6 and 9.5 gcm⁻² shielding depths, respectively, in the main stack of the P0006 experiment on LDEF.

Experimental measurements are compared with the model calculations of Armstrong and Colborn (1992) in Fig. 22. This comparison agrees well at 25 MeV, which may reflect the small contribution of secondary particles under thin shielding. At 114 MeV the experimental value is significantly higher than the model calculation. This requires further analysis of the contribution of secondary particles at this depth. Differences may also arise from the fact that (probably) not identical solid angle intervals were used to measure and calculate fluences. These difficulties in the comparisons would disappear if calculation of directional distribution of stopping proton volume density were available. Concerning the observed high anisotropy of trapped protons; a measurement with better statistical power would also be reasonable for comparison of experimental results with model calculations.

Figure 22. Comparison of calculated LDEF mission fluence of eastward directed trapped protons (filled circles interconnected with lines) (Armstrong and Colborn, 1992) with the upper limiting results of our measurements (filled circles with error bars).

4.1.3. Relativistic Fe Ion Measurements

The directional distribution of galactic cosmic ray (GCR) Fe particles were measured using two sheets of CR-39 etched track detectors from the center of the main stack (under 6.5 gcm^{-2} shielding) and from the four side stacks (under 1.3 gcm^{-2} shielding). The CR-39 sheets were etched in 6.25N NaOH solution at 50°C for 36 hours. The central area of the CR-39 doublets was scanned for tracks in coincidence at the two adjacent surfaces. Track pairs were further examined to determine if there were corresponding tracks at the top surface of the upper sheet and the bottom surface of the subsequent sheet (four surface particle events). The four surface tracks were then selected on the basis of equal sizes of the four aligned tracks, demonstrated that the particles passed through the CR-39 doublet with high (relativistic) energy.

Figure 23. Arriving directions of galactic cosmic ray Fe particles with relativistic energies looking to west direction from the center of the P0006 experiment on LDEF. Particles with dip angle greater than 40° could not be measured because of the critical angle cutoff of the detector.

I have compared the differential LET spectra of four surface tracks found in LDEF detectors to a differential LET spectrum of similar tracks measured on a detector flown on the Shuttle in an orbit close to that of LDEF (STS-32). I have found that practically all the four surface tracks, measured in the LDEF sheets, must be produced by relativistic Fe ions. Nonrelativistic GCR ions are mainly excluded by the geomagnetic cutoff in a 28.5° orbit. In the case of the Shuttle flight detector other than Fe galactic cosmic rays are also included in the differential LET spectrum, but the peak caused by the Fe particles is clearly defined. The greater abundance of Fe ions relative to neighboring elements is evident, and the position of this peak in the differential sensitivity (reduced etch rate ratio) spectra can be used for internal calibration of the detector material. In the LDEF detectors the measured tracks also formed a peak in the differential sensitivity spectrum. The position of the peak of the measured particle tracks appeared at the same sensitivity as the peak of Fe ions in the case of the Shuttle spectrum. This is very strong evidence that these four surface event were caused by relativistic

GCR Fe ions. Furthermore, in the case of the LDEF detectors GCR ions, which are lighter than Fe could not be measured, because their LET was too small to be detected due to the short etching time. (The short etching time for LDEF samples were made necessary by the high total track densities.)

Figure 24. Arriving directions of galactic cosmic ray Fe particles with relativistic energies in the side stacks of the P0006 experiment on LDEF. The empty circles show the arriving directions and the filled circles show the departing directions.

A total of 300 particles in the P0006 main stack and 100-100 particles in each of the P0006 side stacks were identified as relativistic GCR Fe ions. Most of the tedious work of scanning and measuring of sizes and positions of these tracks was done by Luis A. Frigo. From the positions of the tracks of the same particle on the different detector surfaces I have determined the azimuth and dip angle of each particle. Since the moving direction of these high-energy particles along the trajectories could not be determined, I have assumed that all the particles arrived from the west and moved toward the east both in the case of the main and in the side

stacks. The reason for this is that the P0006 experiment was on the west side of the satellite, and the aluminum equivalent shielding of the P0006 experiment from the east directions was in the order of 40 to 150 gcm^{-2} (Colborn and Armstrong, 1996), which is large enough to stop even these high-energy particles.

The directional distribution of GCR Fe particles arriving at the center of the main stack at a depth of 6.5 gcm^{-2} is given in Fig. 23. No particles were detected with polar angle θ (measured from the normal to the detector sheet, where $\theta = 90^\circ - \delta$ and δ is the dip angle) greater than about 40° because of the dip angle cutoff of the CR-39 etched track detector. Scanning efficiency was also less than 100% for particles incident near the dip angle cutoff, because of the difficulty in finding the corresponding tracks on the first and fourth surfaces of the CR-39 doublet. Particles arriving from the west are found to be close to isotropic in directionality but with some extra particles arriving from the south-west parallel to the Earth's surface.

The directional distributions of Fe particles arriving at the four side stacks, under 1.3 gcm^{-2} shielding are shown in Fig. 24. The empty circles show the arriving directions and the filled circles show the departing directions. The shielding of the LDEF was to the east side and there were shieldings of the P0004 'Seeds in Space' canisters extended from this direction to about 10° to the west (slightly over to the center of the graphs). Results can be interpreted as arriving particles from the west in one side stack are from the same population as departing particles (towards east) are in the side stack at the opposite side of the main stack. The observable slight anisotropies, especially in the case of side stacks B and D, strongly support this hypothesis.

4.2. Radon Concentration Measurements

4.2.1. Radon Measurements in Houses

In Hungary radon concentration measurements in houses started in the second half of the 1960s (Tóth, 1977; 1983). Radon concentration measurements in caves with etched track radon monitors were started in the Institute of Nuclear Research by G. Somogyi *et al.* in the 1970s (Somogyi *et al.*, 1983). In these years etched track detector radon measurements were also extensively done in uranium exploration in collaboration with the Mecsek Ore Mining Corporation (Somogyi *et al.*, 1982). Measurements in houses were done in collaboration with the Public Health Institute of County Borsod (Paripás *et al.*, 1984; Déri *et al.*, 1992) and in collaboration with I. Nikl from the “Frédéric Joliot-Curie” National Research Institute for Radiobiology and Radiohygiene, (Somogyi *et al.*, 1989). In the 1980s and 1990s many other organizations (mostly universities and high schools) started radon measurements in Hungary. I. Nikl (1995) has conducted a national survey of indoor radon concentrations in about 1000 houses with electret detectors. However, as of today, probably the largest number of indoor radon concentration measurements was done by the group of the Lauder School (Budapest) headed by E. Tóth. In the 1990s they have measured radon concentration in the order of 10 000 rural houses searching for houses with elevated radon levels. Most recently significant efforts were also made at the University of Veszprém to do radon research in a radon prone area.

A small scale national survey

In a joint research program of the “Frédéric Joliot-Curie” National Research Institute for Radiobiology and Radiohygiene (NRIRR) and of the Institute of Nuclear Research in 1985-1987 we have made a small scale survey study of indoor radon concentration in Hungarian dwellings. The measurements were organized by I. Nikl (NRIRR); done by our group; and I have made the data evaluation. Results are published in Somogyi *et al.*, (1989) and also in Csige (1991a).

There were 122 houses selected, each house associated with a meteorological station of the National Meteorological Service of Hungary. Although the selection of the houses could be considered to be randomly done, it has not represented the full Hungarian building stock.

Radon concentration was measured in the most frequently used room of the houses using the inverted diffusion cup technique. The radon monitors were changed in approximately every three months. Exposure intervals were selected to match with the seasons of the years. The survey started in 1985 and ended in 1987. There were altogether 10 series of measurements and 1220 radon concentration data obtained.

I have calculated the average radon activity concentration for each house from the 10 measured values. The distribution of these data shows a lognormal distribution with a geometric mean (GM) of 47.5 Bqm^{-3} and geometric standard deviation (GSD) of 1.72 (Fig. 25). This result means for example that 66% of the houses are expected to have average radon activity concentration in the 28-82 Bqm^{-3} interval. Arithmetical mean was found to be 55.2

Bq m^{-3} with arithmetical standard deviation of 38.0 Bq m^{-3} . Our result is in the order (or may be slightly higher than) that was found in most countries surveyed in Europe and in America (ICRP-50). Annual effective dose for a person associated with this arithmetic mean value is around 0.8 mSv (dose conversion according to ICRP-65).

In many individual houses I have found significant seasonal variation of indoor radon concentration with typically winter maximum and summer minimum. The same, however, cannot be easily said about the GM of the 122 houses (Fig. 26). This average varies between a factor of two except the data for the period between December 1986 and February 1987, which has a significantly higher value. This later period was an extremely cold winter in Hungary, which might have got residents to reduce ventilation in their homes in order to keep warm. (Error bars on Fig. 26. show intervals determined by 1 GSD and not the accuracy of measurement.)

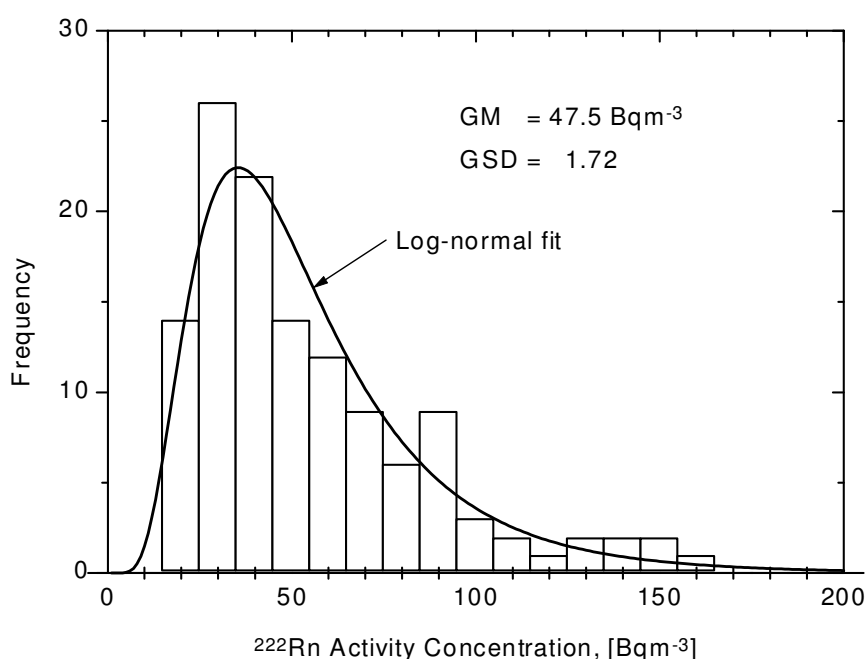


Figure 25. Distribution of average radon activity concentration in 122 houses.

I have found significant difference between arithmetical mean values of indoor radon concentration in houses built from adobe (74 Bq m^{-3}) and brick material (37 Bq m^{-3}). Houses built from brick were further classified as the measurements were done on the ground floor or on the first floor. I have found significant difference between these two cases with mean values being 42 Bq m^{-3} in ground floor and 24 Bq m^{-3} on the first floor, respectively. This suggests that main source of indoor radon is the soil in the ground floor of these houses. Considering the also significant difference between the mean values of indoor radon concentrations in the ground floor of brick (42 Bq m^{-3}) and adobe (74 Bq m^{-3}) houses, the role of the building material as significant source of indoor radon in adobe houses is also likely.

In the following I will compare these results to some recent measurements done in Hungarian houses by others. In 1993-1994, I. Nikl and coworkers have done a more wide national survey of indoor radon concentrations in 998 Hungarian dwellings. Houses were selected to cover the

whole area of the country uniformly and, at the same time, to represent the full Hungarian housing stock. They have used electret radon detectors for measurements (E-Perm, Radon Elec. Inc. USA). Measurements lasted over 1 year in each house. I. Nikl (1995) has found that the arithmetical mean radon activity concentration in these houses was 129 Bq m^{-3} . The population weighted average radon activity concentration was 107 Bq m^{-3} . Both of these values are about a factor of two higher than we have obtained in our above discussed small-scale survey and they compare well with averages found in countries known to have serious radon problem (Sweden, Finland for example).

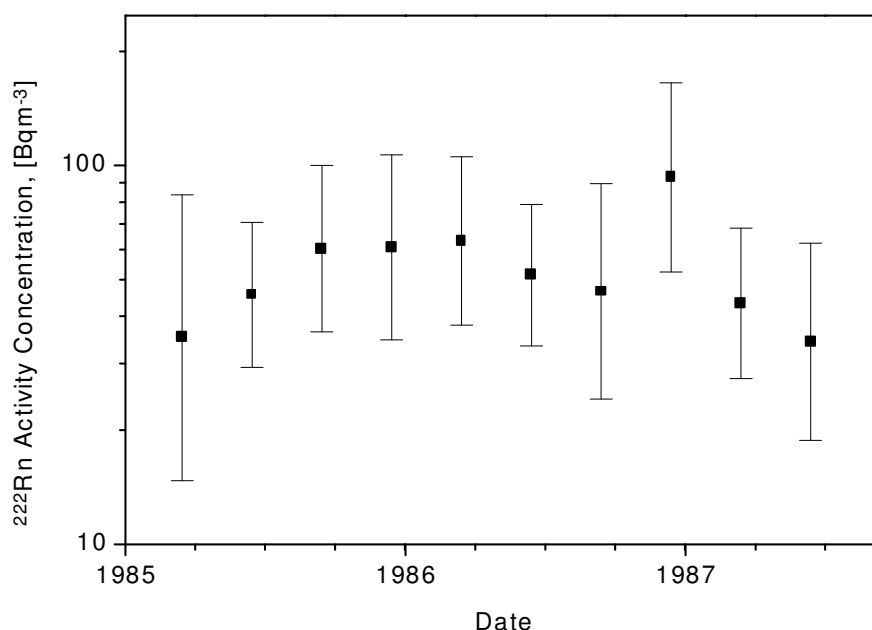


Figure 26. Temporal variation of GM radon activity concentration of 122 houses. Error bars represent intervals determined by 1 GSD not the accuracy of measurement.

E. Tóth *et al.* (1996) have also reported that arithmetical average of indoor radon concentrations in more than 6000 rural houses was found to be around 120 Bq m^{-3} . They have used etched track detectors to measure radon concentration normally nearly for 1 year in each house. It is generally believed that rural houses on average must have significantly higher indoor radon concentration than national average. Therefore, according to their results, the national average radon concentration in Hungarian dwellings is expected to be significantly less than 120 Bq m^{-3} .

The reason(s) for these relatively large discrepancies in the discussed surveys has not yet been resolved. This means there is a relatively large, i. e. at least a factor of about 2, uncertainty in the estimate of the average radon concentration in Hungarian dwellings. Therefore, as currently ongoing measurements do not address this question, there will remain some doubts for a while what the actual size of the “radon problem” in Hungary really is.

A case-control study of radon and lung cancer in eastern Hungary

In a joint research program of the Public Health Institute of County Borsod and of the Institute of Nuclear Research in 1989-1991 we have made a case-control study of radon and lung cancer in dwellings. The goal of the study was to adapt a complex survey technique and to apply it for investigation of the possible relation between indoor radon concentration level and lung cancer in a village where a high (526 ± 44 cases per million per year is the average of the last 8 years) lung cancer incidence with relatively low deviation is shown. Zs. Déri at the Public Health Institute of County Borsod designed and conducted the survey. The radon concentration measurements, however, were done completely in our laboratory. Results are reported in Déri *et al.*, (1992) and they are shortly summarized here.

There were 60 houses in the case group and 140 in the control group. The members of the case group were selected on the basis of data of the local oncological institute and the individuals of the control group were the employees of the Public Health Institute of the town investigated and their relations and friends. The radon concentration was measured in dwellings and in the soil gas of the nearby gardens of the selected case and control group houses using the inverted diffusion cup technique. One radon monitor was placed in the most frequently used room of the house for 1 year and another one was dug in the garden to about 0.4 m depth and covered by soil for 3 to 4 months during the winter season.

No correlation was found between the measured radon activity concentration in the houses and in the nearby soil. Significant difference was found between mean values of indoor radon concentration in houses with cellars (105 Bqm^{-3}) and without cellars (199 Bqm^{-3}). A weaker but still significant difference was found between mean values of indoor radon concentration in houses built from adobe (199 Bqm^{-3}) and other materials (149 Bqm^{-3}). It was concluded that indoor radon concentration depends dominantly on the building mode of houses (presence or lack of a cellar) and less on the building material.

From the database obtained 33 individuals from the case group and 66 individuals from the control group were selected for evaluation. Data selection was based on arriving at similar distributions of other than radon affecting factor (such as smoking) both in the control and in the case groups.

The arithmetical averages of the measured radon activity concentration were 192 Bqm^{-3} (corresponds to 3 mSv/y effective dose) and 155 Bqm^{-3} (2.3 mSv/y) for the case and the control groups, respectively. The difference was found to be significant at 95% confidence level. This result do not contradicts the supposition that the most important cause of lung cancer, after smoking, may be elevated indoor radon exposures.

4.2.2. Radon Exposures in Caves

People, who go to caves, are taking some risks. One of these (may be not the highest) is due to inhalation of short-lived radon daughters. Typical radon levels in caves are between 1 and 10 kBq m⁻³, about 100 times higher than typical levels in homes. However, people usually do not spend that much time in caves (since the Stone Age). Most people do not go to cave at all. Many others do it occasionally as tourist. Tourists spend in caves very short time, may be a few hours a year. Their effective doses from inhalation of radon daughters in caves are negligible comparing to annual effective doses they receive from other natural sources (including indoor radon). Other groups of people, however, spend longer periods in caves. These groups include workers (tour guides and staff of show caves) and cavers (persons engaged upon caving activities as a recreation). Some caves are used for therapeutic treatment of patients, suffering from asthma. Patients and the staff of the therapy are also concerned taking higher risk. These groups benefit different ways and probably very different amount from going to caves. When making smart decision on going to a cave, risks and benefits are properly balanced. In this section I do not deal with such a balancing, but give estimates of risk due to inhalation of short live radon daughters from the air of some Hungarian caves.

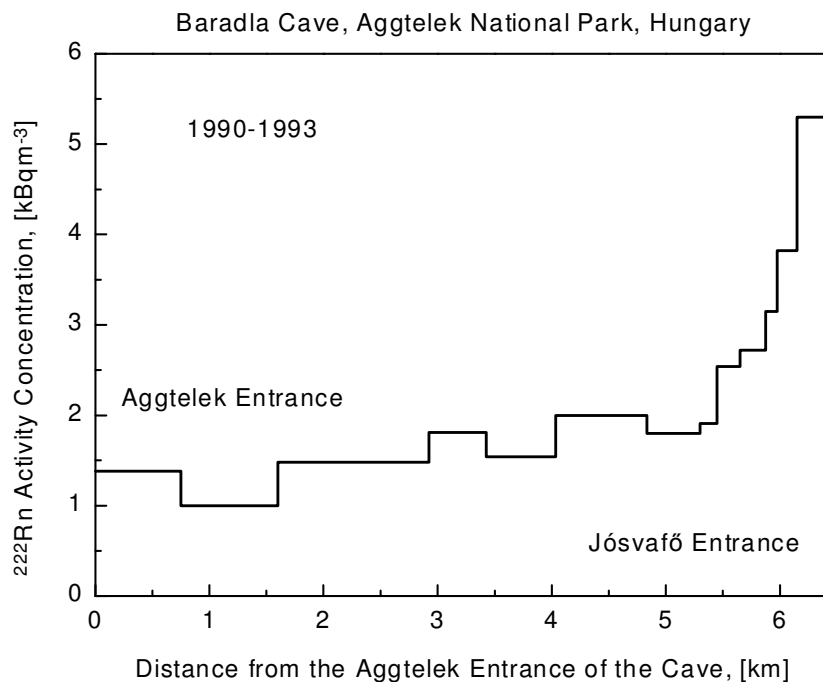


Figure 27. Spatial variation of 4 year average ²²²Rn activity concentration in the Baradla cave as a function of the distance from the Aggtelek entrance of the cave. Measurement points were at the middle of each interval.

Initiated and started with great impetus by G. Somogyi in 1978, up till now we have done regular ²²²Rn measurements in more than 30 Hungarian karstic caves (Hunyadi *et al.*, 1991; Hakl *et al.*, 1992; and Hakl *et al.*, 1997). We used etched track and semiconductor based radon monitors. The etched track technique was used to measure spatial distribution and long-term temporal variation of radon concentration in a given cave. Usually 3-20 measuring sites were established in each cave investigated, spaced equidistantly mostly in the main passages of the caves. The etched track detectors were changed approximately in every 30 days over at least a

3-4 year period. Since 1991 we use Dataqua type "real time" radon monitors to measure fast (hourly, diurnal) temporal variation of radon at some characteristic sites. Some Dataqua instruments were also equipped with air temperature, pressure, relative humidity and water level measuring capabilities to study interrelationships between these parameters.

We have found ^{222}Rn concentration in caves may vary significantly both with space and time. Fig. 27 shows, for example, the spatial variation of time averaged (over the years from 1990 to 1993) ^{222}Rn concentrations in the Baradla cave as a function of the distance from the Aggtelek entrance of the cave. The higher ^{222}Rn activity concentration measured at the Jósvalő entrance part (right side part on Fig. 27) of the cave is caused by the higher ^{226}Ra content of rock and by a special local air circulation pattern (Hakl et al. 1997).

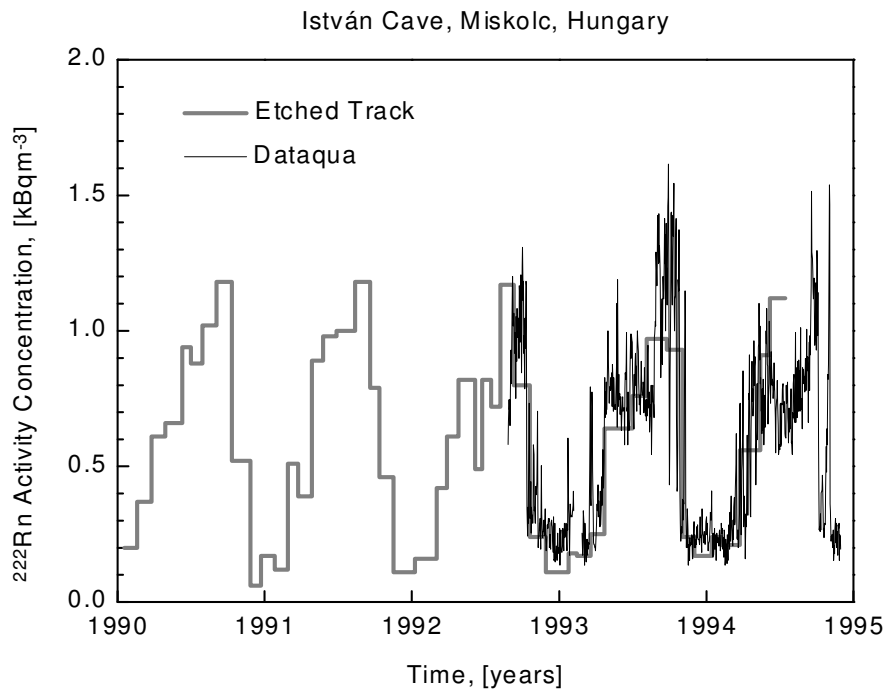


Figure 28. Temporal variation of radon activity concentration at the therapeutic part of the István cave (Miskolc, Hungary).

Fig. 28 shows a seasonal variation of ^{222}Rn concentration in the Szent István cave (Miskolc, Hungary), a feature, we found typical in the main passage of horizontal entrance caves. This type of temporal variation was first explained by Géczy *et al.*, (1989). Chimney effect induced air circulation carries ^{222}Rn from the fracture system to the cave in summer times, while in winter times, fresh air entering the cave through the entrance reduces ^{222}Rn in horizontal cave gallery. One advantage of these type of caves is that time variation of radon concentration in them is fairly well predictable.

In vertical entrance caves, however, atmospheric pressure variation is the dominant control parameter on radon concentration (Hakl *et al.*, 1996c). A substantial pressure drop caused by a deep low-pressure weather system will cause air to flow out of the cracks, carrying radon with it. High-pressure front pushes fresh air into the cave through the entrance and also pushes some air back to the fractures and pores. Radon concentration variation in these caves is

predictable as far as weather fronts can be predicted. The Cserszegtomaj well cave in Hungary is a typical example of this type of caves (Hakl *et al.*, 1996c).

In some caves both temperature and pressure control on incave radon concentration are dominant. An example from the Hospital cave of Tapolca (Hungary) is shown on Fig. 29. From the results of these measurements we may conclude that in order to obtain reliable estimates of radon exposures in caves a sufficiently detailed measurement of the incave ^{222}Rn activity concentration (with spatial and temporal resolution) is needed. Or alternatively the wearing of a personal radon dosimeter is suggested.

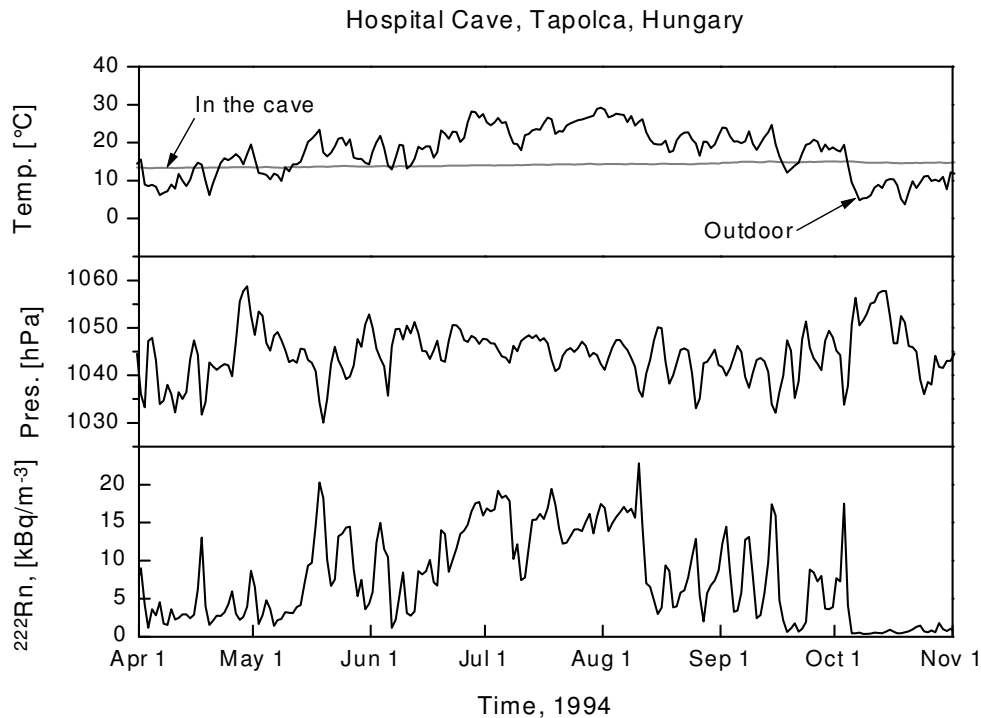


Figure 29. Temporal variation of daily average temperature, pressure and radon activity concentration at the therapeutic part of the Hospital cave (Tapolca, Hungary). (Measurement was done by A. Várhegyi, Mecsekurán Ltd., Pécs, Hungary, using a Dataqua instrument.)

To calculate the effective dose due to inhaled short-lived radon daughters, I have used our radon measurements and detailed records of people who have visited the caves. First, I have used Dataqua radon records, if there were any available. If Dataqua radon records were not available at a given site or time period the nearest etched track radon results were used. To obtain equilibrium equivalent decay product exposure I have used equilibrium factor of $F=0.4$ between radon and its short-lived daughters. I have chosen this value because of the findings on equilibrium factor for example in Postojna cave measured by Butterweck *et al.* (1992) and Jovanovic *et al.* (1992) and in Royal Cave (Australia) measured by Solomon *et al.* (1992). Szerbin (1996) reported similar F values in some Hungarian caves, too. These authors also found unattached fraction of daughters to be relatively high ($f=0.1$) when comparing it to mine or indoor atmospheres ($f=0.01$), leading 1.4-2.5 times higher effective doses in caves. I did not consider this effect in my calculations. Instead, I have used conversion factor between potential alpha energy exposure and effective dose as given in ICRP-65 for members of the public (1 mJm^{-3}h is equivalent to an effective dose of 1.10 mSv).

Recently, we have continuously monitored radon concentration in all five caves in Hungary, where therapeutic treatment of asthma patients is going on. Earlier we have reported effective doses (estimated according to ICRP-50) of patients in one of these caves (Szemlő-hegy) (Hunyadi *et al.* 1995). Later, I have revised these data according to ICRP-65 (1993) and we have published it together with data from the other four therapeutic caves (Csige *et al.* 1996). In the case of the other four therapeutic caves annual effective doses of patients were determined for the year of 1994. Summary of results is presented in table I.

We have also monitored some show caves in Hungary. In the case of the best known of these (Baradla cave), I have estimated annual effective doses for each tour guide for the years between 1990 and 1994. Summary of results is included in table I.

Table I. Annual effective dose, mSv

Cave name	Period	Sample size	Mean (Range)	Geometric mean (Geometric STD)
Abaliget	1994	127 patients	0.54 (0.03-1.26)	0.40 (2.2)
Béke	1994	56 patients	1.91 (1.86-1.97)	1.91 (1.0)
Szemlőhegy*	1990-1992	229 patients	0.85 (0.10-5.00)	0.62 (2.1)
István	1994	360 patients	0.06 (0.01-0.17)	0.04 (2.4)
Tapolca	1994	481 patients	0.87 (0.04-2.19)	0.45 (3.7)
Baradla	1990-1994	12 tour guides	2.66 (0.12-5.55)	2.13 (2.2)

*In the case of Szemlő-hegy cave cumulative effective doses over 1990-1992 are given in mSv.

Based on their internal working time reports, I have found annual effective doses for staff members of therapy are about 0.4 mSv in Abaliget cave, 0.12 mSv in István cave, 6 mSv in Béke cave. Tour guides in Abaliget cave receive about 12 mSv annually.

Cavers and researchers visit caves for very different reasons. Some of them go to the same cave regularly, others visit as many different caves as they can. Most active cavers may spend in caves up to a few hundred hours annually; therefore they may be exposed to higher doses. Using a Radamon personal radon dosimeter, J. Hakl has measured his own annual effective dose for example, while working in different caves, and found it was higher than 30 mSv in 1992. In another case, in 1996, a group of cavers spent a whole week in the Cserszegtomaj well cave, which has the highest ²²²Rn activity concentration known in Hungaria caves, and each of them received an effective dose of 16 mSv.

Although these people go to cave willingly, the order of these doses call for attention. Therefore we have suggested that:

1. all caves should be tested against radon at least for one year before opening to public or therapeutic treatment or used frequently by the same group of people;
2. groups at higher risks should be informed. Their exposure should be limited;
3. personal radon dosimetry is highly recommended for the most active cavers and especially for tour guides and assistants of therapy.

4.2.3. Radon in a Dry Carbon Dioxide Spa

Mátradereske (Mátra Mountain, Hungary) is located in the region of the so-called Darnó line, which is one of the most mobile crustal parts of Hungary (Rumpler and Horvath, 1988). At a depth of about 600 meters beneath the village, the Triassic limestone is covered by Triassic clay-shale, on which Eocene andesite is layered with a thickness of about 400 meters. On the surface, a few meters thick Oligocene clay covers partially the andesite. The fragmented limestone serves as a karstic aquifer. Its water is saturated with gas, containing mostly carbon dioxide (cca. 95%). Carbon isotope ratio study showed that CO₂ has geological origin, and is created by thermal dissociation of the limestone (Hertelendi *et al.*, see in Szilágyi, ed. 1992). The gas moves towards the surface through the faults of the andesite. It escapes to the surface mostly where the Oligocene sediment is thin or missing.

There is a natural, dry carbon dioxide spa in the village, currently run (since 1995) by the City Hall of Mátradereske. The spa is based on an about 2.5 m deep pit dug into the soil at the area most heavily affected by the carbon dioxide exhalation. Carbon dioxide enters the pit from the walls of the pit and from a 20 m deep bore hole drilled into the soil at a few meters distance from the pit. The spa is housed in a wooden building, which also includes a resting room and a hygienic block. The treating room, where the pit is, has many openings to the outdoor environment at about 0.2 m beneath the floor (0 meter) level of the room. As a result of these openings, the pit is normally filled with carbon dioxide up to this level.

Patients suffering from stenosis visit the spa for curing. Patients can go into and stand or sit in the pit at different levels on a stepped wooden trestle-work. Lower parts (mostly the legs) of their bodies are exposed to carbon dioxide, which gas diffuses into their bodies through the skin. One treatment typically lasts for 20 minutes. A cure consists of 18 treatments over a 3 weeks period. Cures are run seasonally, normally from April to October with a break in August.

In addition to the carbon dioxide, there is a relatively high ²²²Rn activity concentration in the spa. During curing periods, we have regularly measured ²²²Rn activity concentration in the spa using Radamon etched track type radon detectors and Dataqua instruments. The Radamons were placed for different locations in the treating room including the pit. The Dataqua instruments were placed either at a corner of the treating room at floor (0 meter) level, or at the wall of the treating room at about +1 meter level. In addition personal radon dosimetry of staff were regularly done using Radamons. In most of the following I will limit the discussion to the results obtained during the period from 15th of April 1996 to 18th of October 1996.

We have found that the ²²²Rn activity concentration in the treating room varies as a function of altitude (Fig. 30). At the lowest step level (-1.2 meter) in the pit it is around 140 kBqm⁻³, while at the level of the mouth (at around +0.4 meter) of a patient, who is standing in the lowest step, it is around 2 kBqm⁻³. At floor (0 meter) level it was around 4 kBqm⁻³.

An undisturbed one week long temporal variation of ²²²Rn activity concentration at floor level in the treating room of the spa is shown on Fig. 31.

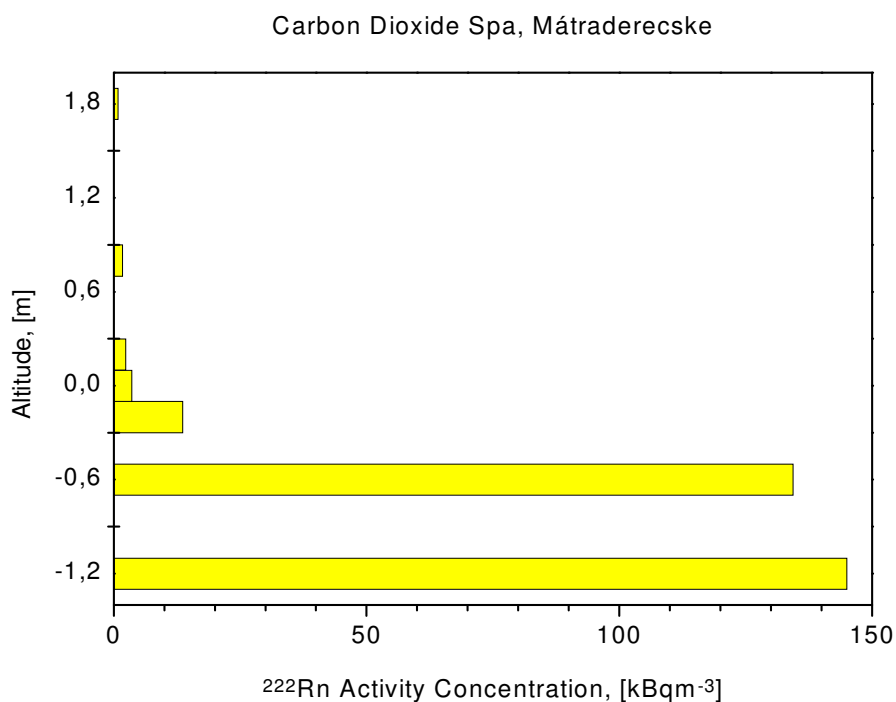


Figure 30. Variation of ^{222}Rn activity concentration in the treating room of the carbon dioxide spa as a function of altitude. 0 meter corresponds to floor level. Patients usually stand on the lowest step at -1.2 meter altitude of their legs.

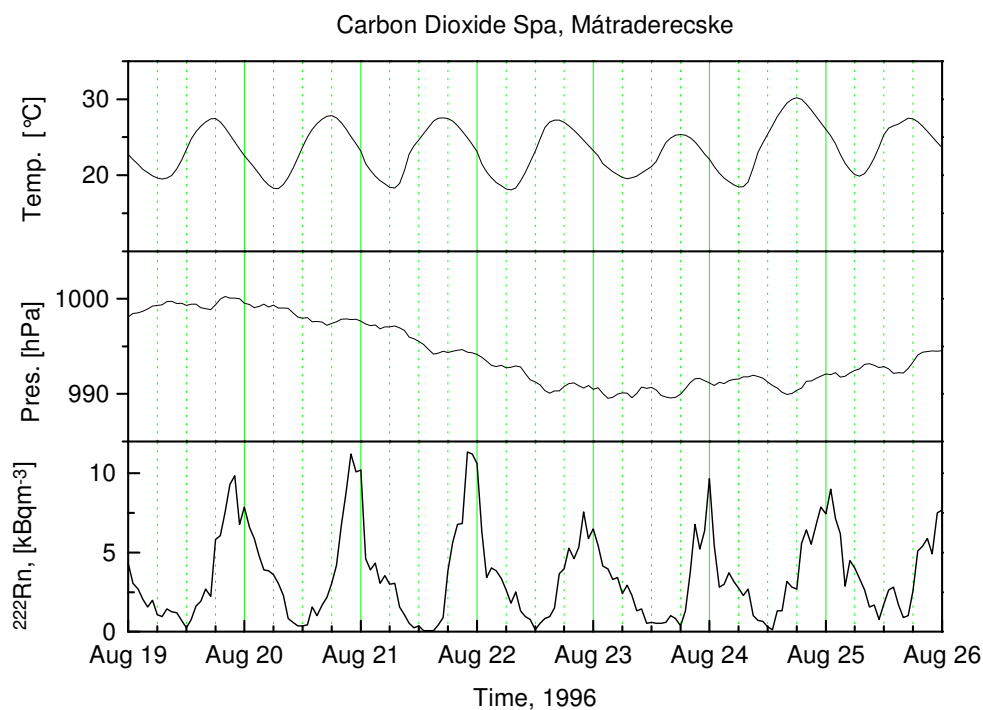


Figure 31. A typical undisturbed variation of temperature, pressure and ^{222}Rn activity concentration as a function of time at floor (0 meter) level in the treating room of the spa. There was no therapy in the spa during this week.

I have found that the ^{222}Rn activity concentration at 0 meter level shows a regular diurnal variation. It has a minimum usually less than 2 kBqm^{-3} at around noon, and a maximum of about 5 to 10 kBqm^{-3} at around 22 o'clock in the evening. Concerning schedule of treatments this type of diurnal variation of ^{222}Rn activity concentration is considered to be lucky. Fig. 32 for example shows the average daily variation of ^{222}Rn activity concentration on Tuesdays for three different periods. In the first (from 15th of April to 14th of July 1996) and in the third (from 21st of August to 18th of October 1996) periods there was therapy in the spa in the afternoon hours on Tuesdays, while in the second (from 15th of July to 20th of August 1996) period there was not. Fig. 32 shows that the ^{222}Rn activity concentration at 0 meter level is increased by a factor of 2 during treatment hours. This effect may be due to the disturbing mixing of gases in the spa by the patients.

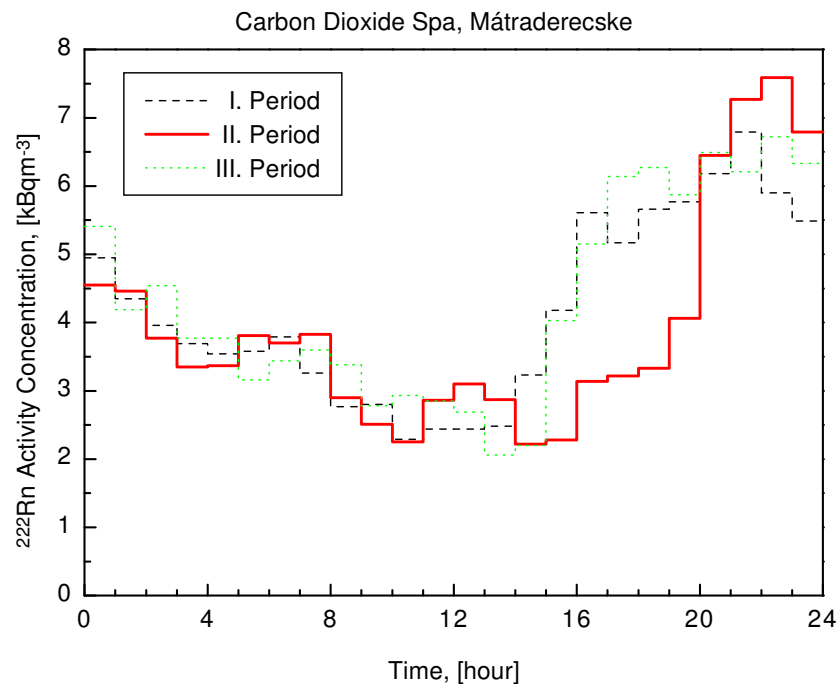


Figure 32. Average daily variation of ^{222}Rn activity concentration in treating room at floor level on Tuesdays for three different periods. In the II. period there was no treatment in the spa. In the other two periods treatments were done in the afternoon hours.

I have estimated the effective doses due to inhalation of ^{222}Rn and its decay products for the patients and for the staff of the therapy. In addition I have also estimated the effective dose due to ^{222}Rn that diffuses into the body through the skin of the patients. I thought this radiation pathway may contribute significantly to effective dose, because radon concentration at the legs of the patients is about two order of magnitude higher than at the mouth level. It is known (ICRP-65) that the contribution of effective dose due to inhaled ^{222}Rn only is about 2% of inhaled decay products. First, I have thought the same conversion factor may apply to the ^{222}Rn entry through the skin as is used for inhaled ^{222}Rn . Then, in order to obtain a more accurate dose conversion factor for this radiation path way, I have used a compartment model to describe the transport of ^{222}Rn in the human body. This model is based on the paper of Sharma et al. (1996) and references in it. Transfer coefficients and tissue weighting factors were also taken from ICRP-60 and ICRP-65. I have found that the dose conversion factor for

^{222}Rn that enters the body through the skin is $0.002 \text{ nSv Bq}^{-1}\text{m}^3\text{h}^{-1}$, about 35 times smaller than the one from inhalation of ^{222}Rn only ($0.07 \text{ nSv Bq}^{-1}\text{m}^3\text{h}^{-1}$). This is because much of the ^{222}Rn that enters the body through the skin, first is collected in the veins, then it is exhaled before blood in the arteries would transport it to other organs. Even though, the contribution of this component to total effective dose of patients in the spa is significant, it is about 4%. Total effective dose of patients due to inhalation of ^{222}Rn and its decay products and from diffusion of ^{222}Rn into their body is about $30 \mu\text{Sv}$ for one treatment and about 0.6 mSv for a cure.

Predicted uncertainty of the above dose conversion factor is about a factor of 8. Currently, it has no experimental verification. Prediction of ^{222}Rn content of air exhaled by a patient, who is exposed to 140 kBqm^{-3} ^{222}Rn activity concentration for 15 minutes at the skin only, as a function of time is shown on Fig. 33. This prediction can be tested against measurements.

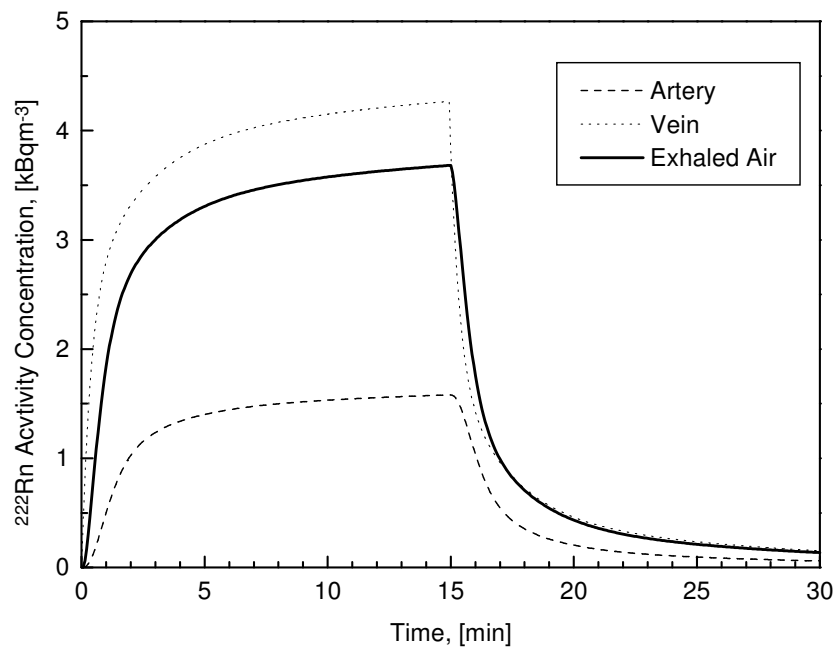


Figure 33. Result of compartment model calculation. Variation of ^{222}Rn activity concentration in the blood and in the exhaled air as a function of time in the case, when skin surface of the body is exposed to 140 kBqm^{-3} ^{222}Rn activity concentration for 15 minutes. ^{222}Rn content of inhaled air was assumed to be zero.

Personal effective doses of staff of the therapy due to inhalation of ^{222}Rn and its decay products were estimated from personal radon dosimetric results. The staff carried the Radamon detectors as a medallion on a necklet, as close as possible to their mouth. They wearied it during working hours in the spa, otherwise the detectors were stored in the resting room, where an additional Radamon measured the background ^{222}Rn activity concentration. Effective doses of staff ranged from 0.9 mSv to 4.2 mSv for the whole season. Highest dose was received by a safeguard who spent much of his time in the treating room paying attention on patients in the pit.

4.3. Future Perspectives in Radon Studies

Efficient search strategies for radon-prone areas as well as efficient mitigation techniques require deep level of understanding of radon occurrence in nature, its origin, its transport in soil and its entry into houses. Influencing geologic factors, soil properties, weather parameters, house characteristics and inhabitant behaviors make the problem very complex. The scientific aim of one of our recent project is to develop criteria for building sites and for indoor radon concentration, when all relevant influencing factors are taken into account. The practical aim of the work is to develop a general physical, mathematical and computer model to describe the transport of radon in soils and its entry into houses. The improvement of such models is also crucial to increase effectiveness of radon tracer techniques. It is also the aim of the program to make control measures at selected “radon test-houses” to validate model calculations.

Much of the work in this field we do in international collaboration with partners from France, Germany, Sweden, and Spain. Their work is supported by the EU Human Capital and Mobility program. Our work in Hungary is supported by the Hungarian National Scientific Research Fund. These supports is kindly acknowledged here.

There are many radon transport models developed. Some of them are very complex (for example Andersen, 1992; Nielson *et al.*, 1994; Ferry *et al.*, 1996), some others “suffer” from oversimplification studying the effect of only one or several factors. In the following I will present some very promising results of model calculations I have elaborated in last years (Csige *et al.*, 1997b). Calculations are compared with results of field measurements.

I limited my study to the non-saturated zone, i.e. from the ground-water table to the surface. Soil is considered to be an isotropic, porous medium with respect to the relevant transport parameters (porosity, permeability, heat conductivity and radon diffusivity). Water saturation was allowed to vary as a function of space, but not as a function of time. Consequently, all transport parameters also varied as a function of space (inhomogenous soil). At the groundwater level I have assumed a constant flux density of geo-gas up-flow. At the soil-air interface the temperature and atmospheric pressure were prescribed as time varying boundary conditions. I have calculated the transport of heat, of soil gas and of radon in the soil solving numerically the appropriate transport equations (Csige *et al.*, 1997b).

We have chosen our radon test-house in Mátraderecske (Mátra Mountain, Hungary). I have also compared my calculations with measurements done at the Lamalou (near Montpellier in South France) radon test-site. Their test-house was chosen on a heavily fragmented karstic terrain, which has a slope around 5°.

We have measured the radon concentration at different depths in the soil with single and multi-channel Dataqua instruments. Measurements were done in the flour-garden at about 1 meter distance from the wall of the test-house. The Dataqua probes were completely buried in the soil. Measurements were done over a whole year with 1 hour reading cycles.

For heavily wet soils, in which the diffusion length of radon $(D_a/\lambda)^{1/2}$ becomes comparable to the sizes of the measuring volume of the probe, the radon concentration in the sensitive volume of Dataqua may significantly lower, than in the pore space (Tanner, 1991). Therefore we limited our attention first to relatively dry periods.

Measured radon activity concentration in soil at 1 m depth was compared to calculated values. This time, I have used a 1 dimensional model. Water content of soil was fitted to values measured in the upper 1 meter thick layer, and at greater depths it increased linearly as a function of depth (saturated at the ground water table level). Gas-flux at the ground-water table was set to zero. Measured time series of atmospheric pressure, and an assumed time series of surface temperature were used as time varying boundary conditions.

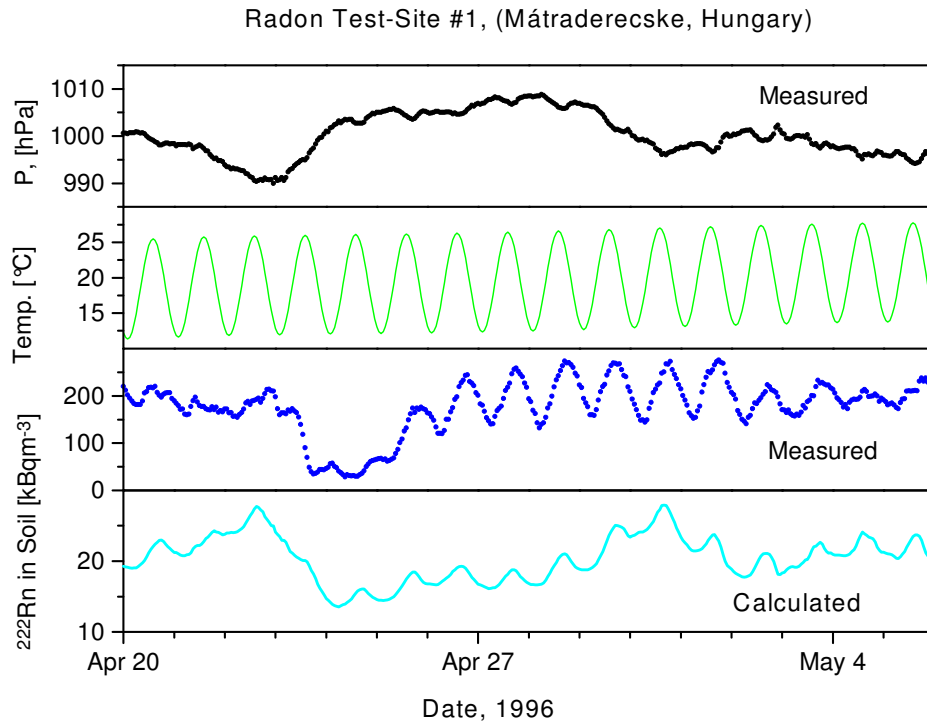


Figure 34. Variation of atmospheric pressure and surface temperature (upper part), and measured and calculated radon concentration in soil at 1 m depth (lower part), as a function of time at the Mátraderecske (Hungary) radon test-site.

The results are presented on Fig. 34. This graph suggests that heat transport in the upper soil layers may have measurable effects on soil radon content. However, the amplitude of the effect is much smaller in the model calculation than in the experiment. Also the average radon level is significantly (by an order of magnitude) higher in the experiment than in the calculation. On the other hand the daily variation, which is related to the temperature effect, is in positive correlation in the measurement and in the calculation.

This was not always the case, however. I have made another running of this program when the experimental input parameters of the test site were changed to the ones measured at the Lamalou test-site. I have found that in this later case the calculated and measured radon time series varied in anticorrelation with each other (Fig. 35). From this discrepancy I have concluded that the physical model underlying the way surface temperature may effect radon concentration in soil should be fundamentally different in the case of the Mátraderecske and the Lamalou test-sites. At the Mátraderecske test-site an increase in the surface temperature propagates into the top layers of the soil and the soil gas expands a bit. This induces some upward directed gas motion in the soil bringing up some radon from greater depths. Also variation in radon partition coefficient with temperature increases radon in soil air at daytime. (These effects were included in the calculation.)

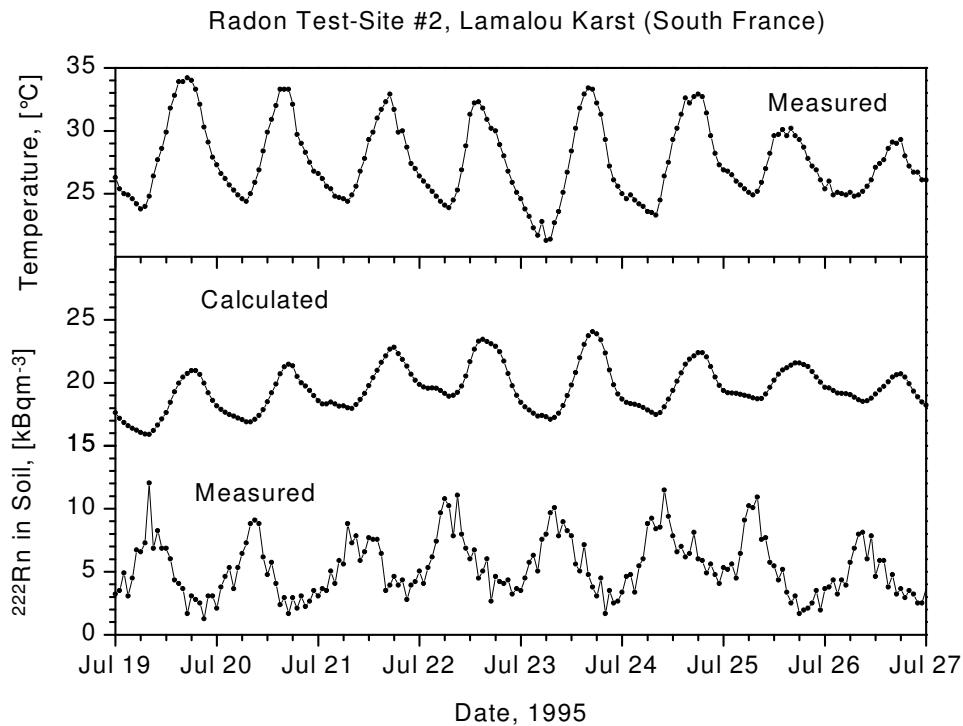


Figure 35. Variation of surface temperature (upper curve), and measured and calculated radon concentration in soil at 1 m depth (lower part) as a function of time at the Lamalou (near Montpellier, South France) radon test-site. (Measurements were done by H. Climent *et al.*, University of Montpellier, France)

In the case of the Lamalou test-site, however, these effects may be far depressed by a more drastic temperature induced effect, which is the air circulation of the karst governed by the chimney effect. In summer time "chimney forces" induce atmospheric air to enter the karst from the terrain, move downwards and leave the karst at lower altitudes, preferable at horizontal cave entrances. At daytime, when the temperature difference between the surface and in the karst air is higher, air circulation in the karst is also intensified. In the night, however, as the surface air cools down, air circulation in the karst slows down, too, allowing more radon to diffuse from deeper to near surface layers. Chimney effect forces were not included in my calculations.

With these two examples, I just wanted to demonstrate the complexity of the problem of radon transport in soils. From these results I have concluded that in order to obtain better agreement between calculations and measurements the physical-geophysical part of the model needed to be more general.

Another aspect of radon studies in soil is to find the best parameters to measure, which give the most cost effective information on the radon source strength of the soil. I have found that perhaps one of the most useful parameter could be the measurement of the ²²²Rn exhalation rate of soil surfaces. The annual average ²²²Rn exhalation rate of a building site would probably be an excellent index for radon source potential. Direct measurement of exhalation rate, therefore has a particular importance. The effects of influencing factors on the free exhalation rate of ²²²Rn should be known in order to interpret measurements properly. The method of exhalation rate measurement can also be critical.

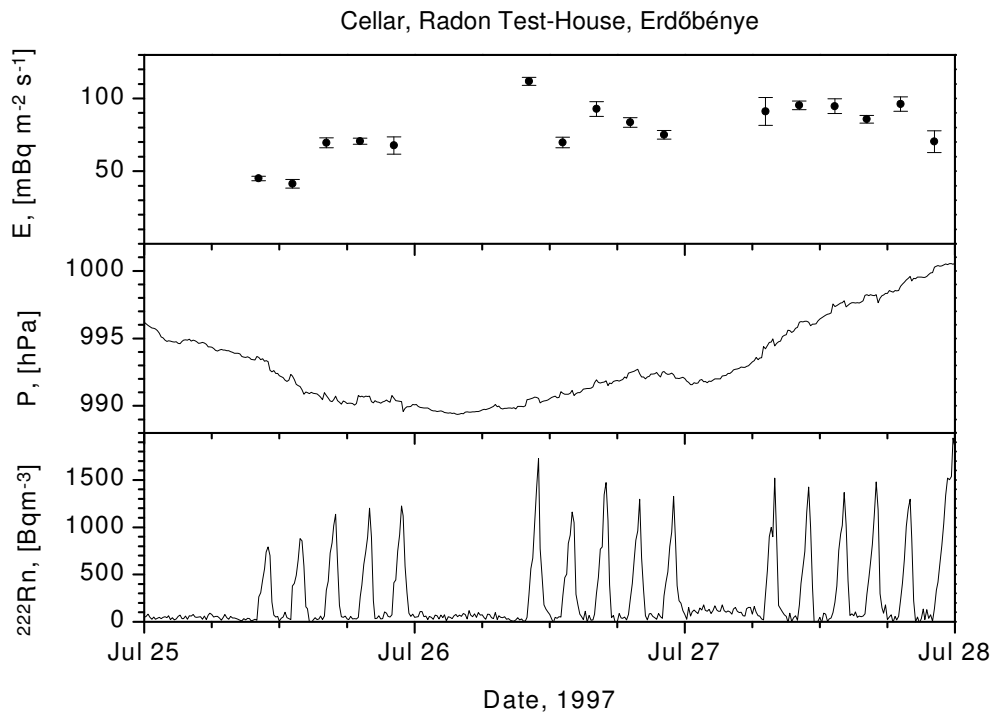


Figure 36. Variation of ^{222}Rn exhalation rate on the floor of a cellar (upper part of the graph) and atmospheric pressure (middle part) as a function of time. Lower part of the graph shows the time series of ^{222}Rn activity concentration as it was measured by an AlphaGUARD ionization chamber. Peaks correspond to 1 hour long ^{222}Rn exhalation rate measurements, which were followed by 2 hours long relaxation periods in outdoor environment.

On Fig. 36 I show results of ^{222}Rn exhalation rate measurements done on the floor of a cellar at three subsequent days using an AlphaGUARD and the method described in the 3.2.3 section of this dissertation. The ^{222}Rn profile in the upper 1 meter layer beneath the sampling spot was monitored by 6 Dataqua instruments. I have found, that while the ^{222}Rn profile in the soil did not change within the accuracy of the Dataqua instruments (about 5% in this case) during these days, the measured exhalation rate showed a fluctuation within a factor of about 2 with seemingly no correlation with weather parameters. Therefore, I consider this fluctuation as due to uncertainty of the measurement technique. On the other hand, a factor of 2 uncertainty should not be considered as too bad in this case. Accuracy of other frequently measured parameters in radon source potential determinations, such as of permeability of soils is usually less accurate than that. And they provide far less direct information on radon source strength of soils than do exhalation rate measurements. Of course the more parameters are measured the more accurate the final radon source potential estimate can be. But we are talking about cost effectiveness here.

In the future there is hope. Even if it turns out that ^{222}Rn is as dangerous as we think today, that there are thousands' life at danger, it can be understood and controlled. We can envisage a safer radiation environment by setting standards (or defining the goals), for example. Then it will probably cost a lot of brain work and field work, and money, too, to achieve it. But the lives saved may make worth of the efforts.

5. Summary

In this Ph.D. dissertation I have presented some results that I have got in the fields of space and radon radiation protection measurements during the past decade while I was working in the Track Detector (later renamed as Radon Group) of the Institute of Nuclear Research (Debrecen, Hungary) and in 1991 and 1992 in the Physics Research Laboratory of the University of San Francisco (San Francisco, California, United States). In the following I have grouped the main findings of the dissertation into three points.

1. *Instrument developments and calibrations.*

I have found that the response of CR-39 to alpha particles can be described by two parameters: the so-called etch induction time and the reduced etch rate ratio (sensitivity), both as a function of particle energy. I have found that, at room temperature, the sensitivity of CR-39 decreases as a function of storage time, even years after its production. On the other hand, I have found that the sensitivity of CR-39 does not depend on the time of the particle incidence, it depends only on the age of the detector at the time of etching.

I have found that irradiation of CR-39 in vacuum increases the etch induction time and reduces the sensitivity significantly if the detector is kept in vacuum at least for a few hours before and for a few minutes after irradiation. I have concluded that dissolved oxygen in the detector material plays an important role in latent track formation.

I have found that post-irradiation sensitization of CR-39 in CO₂ atmosphere increases both as a function of treatment time and as a function of partial pressure of CO₂. This sensitization did not depend on the age of the latent track nor on the energy of alpha particles. I have concluded that CO₂ plays a role in latent track transformation in CR-39.

I have developed and calibrated a passive, etched track type ²²²Rn and ²²⁰Rn concentration measuring detector, called Radamon. I have participated in two international intercomparisons and found that Radamon provided results within 7% of the nominal ²²²Rn exposures reported by the organizers.

I have developed the multiple-etches technique to measure the high linear energy transfer (LET) portion of the LET spectra in CR-39 detector sheets that were exposed on the so-called Long Duration Exposure Facility (LDEF) satellite. I have developed the track anti-coincidence method to measure the mission fluence and directionality of arriving protons in an experiment on the LDEF satellite.

2. *Space radiation measurements.*

I have identified short range high-LET secondary particles, produced in interactions between trapped protons and the C and O nuclei of the experimental detectors, as a major contributor to the LET spectra for low Earth (LDEF type) orbits.

I have found that trapped protons collected in LDEF CR-39 detectors show strong directionality with more protons arrived from the west direction than from any surrounding

directions. I have found that upper limit of mission fluence of trapped protons from the west direction is 6.3×10^6 at 25 MeV and 9.2×10^6 at 114 MeV proton energy.

I have found that directionality of LDEF mission fluence of relativistic Fe ions was slightly unisotropic with some extra particles coming from the south-west direction parallel with the surface of the Earth.

3. Radon concentration measurements.

From the analysis of indoor radon concentration data from a small scale national survey of 122 dwellings in 1985-1987 I have found that the arithmetic average radon activity concentration in Hungarian dwellings is 55 Bqm^{-3} . This value is about a factor of 2 smaller than that was obtained in a larger scale national survey of 998 dwellings done by I. Nikl and coworkers in 1993-1994. However, I have also found that it may be closer to the value that could be expected when comparing to the average of more than 6000 radon concentration measurements done by E. Tóth and coworkers in rural houses. I have concluded that an uncertainty of a factor of about 2 remains in the estimate of the average radon activity concentration in Hungarian dwellings.

I have found that annual effective dose received by tour guides of show caves, assistants of therapeutic caves and by the most active cavers from inhalation of radon decay products in cave air is in the order of 1 to 10 mSv, but significantly higher values may also occur. Because of the order of these doses and due to the known significant spatial and temporal (seasonal, weather front linked, and sometimes diurnal) variation of cave radon concentration I propose personal radon dosimetry for the most active cavers, tour guides and assistants of therapy. I also suggest to test all caves against radon at least for one year before opening to public or therapy or used otherwise by the same group of people.

I have found that the annual effective dose of staff of a dry carbon dioxide spa due to radon and its decay products, is a few mSv. I have found that the contribution of radon, that diffuses into the body through the skin, to the total effective dose due to radon and its decay products is significant for patient standing in the spa. Average cure effective dose of patients, however, is relatively low, it is around 1 mSv.

As a perspective of future radon studies, currently I have been working on a physical-mathematical model to describe the transport of heat, water, soil gas, and radon in the non-saturated zone of inhomogenous soils, and the entry of radon into houses when all relevant influencing (geological, meteorological) factors are considered. My preliminary calculations agreed relatively well (at least qualitative) with experimental measurements that we have done at our Mátraderecske and Erdőbénye (Hungary) radon test-sites. They do not agree, however, at all with measurements done by our collaborating partners at another radon test-site at Lamalou karst terrain (in southern France). I have concluded that in order to obtain better agreement between calculations and measurements the physical-geophysical part of the model should be more general.

6. Summary

In this PhD dissertation I have presented some results that I have got in the fields of space radiation and radon dosimetric measurements during the past decade while I was working in the Track Detector and later Radon Group of the Institute of Nuclear Reserach (Debrecen, Hungary) and in 1991 and 1992 in the Physics Research Laboratory of the University of San Francisco (San Francisco, California, United States). In the following I have grouped the main findings of the dissertation into three thesis points.

1. *Instrument developments and calibrations.* I have calibrated and measured the effects of vacuum and CO₂ on the sensitivity of CR-39 etched track detector. I have found that the response of CR-39 to alpha particles can be described by the so-called etch induction time and reduced etch rate ratio (sensitivity) as a function of particle energy. I have found that irradiation of CR-39 in vacuum reduces its sensitivity significantly. I have concluded that dissolved oxygen in the detector material plays an important role in latent track formation. On the other hand, I have found that post-irradiation treatment of CR-39 in CO₂ atmosphere increases its sensitivity significantly. I have found that this sensitization increased both as a function of post-irradiation treatment time and as a function of partial pressure of CO₂ gas in the treatment vessel. On the other hand, I have found this sensitization did not depend on the age of the latent track and on the linear energy transfer of the particle. I have concluded that CO₂ plays a role in latent track transformation in CR-39.

I have developped and calibrated the Radamon etched track type radon monitor. I have participated in two international intercomparisons of passive radon monitors and found that Radamon provided results within 7% of the nominal radon exposures reported by the organizers. I have developped a method to measure both ²²²Rn and ²²⁰Rn concentration in air with a pair of Radamon radon monitors.

2. *Space radiation measurements.* I have developped the multiple-etches technique to measure the high linear energy transfer (LET) portion of the LET spectra in CR-39 samples that were exposed on the so-called Long Duration Exposure Facility (LDEF) satellite. I have found that the high LET portion of the LET spectra in LDEF samples were significantly higher than pre-recovery estimates predicted. I have identified that the high LET tail of the LET spectra in LDEF samples is mainly consists of secondary particles created in collisions of high energy trapped protons with target C and O nuclei in the detector material.

I have developped the track anti-coincidence method to measure the mission fluence and directionality of arriving protons in the P0006 experiment on the LDEF satellite. I have found that protons show strong directionality with more protons arrived from the west direction than from any surrounding directions. I have found that mission fluence of trapped protons from the west direction was in the order predicted by pre-recovery estimate at 26 MeV proton energy and was may be higher that that at proton energy of 116 MeV.

I have developped a method to identify and measure directionality of relativistic Fe ions in the P0006 experiment on the LDEF satellite. I have found that directionality of mission

fluence of relativistic Fe ions was highly isotropic with some extra particles coming from the west direction parallel with the surface of the Earth.

3. *Radon dosimetric measurements.* From the analysis of indoor radon concentration data from a small scale national survey of 122 dwellings I have found that the arithmetic average radon activity concentration in Hungarian dwellings is 56 Bq/m³. I have found this value is about a factor of 2 smaller than that was obtained in a larger scale national survey by I. Nikl of 998 dwellings. However, I have also found that it may be closer to the value that could be expected when comparing to the average of more than 6000 radon concentration measurements done by E. Tóth et al. in rural houses. I have concluded that an uncertainty of a factor of about 2 remains in the estimate of the average radon activity concentration in Hungarian dwellings.

I have found that staff of show and therapeutic caves receive effective doses from 0.4 to 15 mSv/year from inhalation of radon and its daughters in cave air. I have found that the most active cavers receive effective doses up to about 30 mSv/year. I have concluded protective measures should be taken against radon in the "cave business".

I have developed a physical-mathematical model to describe the transport of heat, soil gas, and radon in the non-saturated zone of an inhomogenous soil in a case, when, at the groundwater table, an upflow of geogas is taking place at a constant rate. I have found that while model calculations agree with experimental measurements at our Mátradereske radon test-site (at least qualitative), they do not agree with measurements done at another radon test-site at the Lamalou karst terrain (in southern France). I have concluded that in order to obtain better agreement between calculations and measurements the physical-geophysical part of the model should be more general.

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