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## **ORGANIZATION**

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Institute of Preventive and Clinical Medicine, Bratislava

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**Session I**

**RADIATION PROTECTION AT WORKPLACE**

**Session II**

**NUCLEAR ACCIDENT:  
MANAGEMENT AND INSTRUMENTATION**

**Session III**

**LOW ACTIVITY IN ENVIRONMENTAL**

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## **RADON IN THE HUMAN LIFE**

***J.Thomas***

*State Office for Nuclear Safety, Praha*

### **Abstract**

Radon causes the utmost but controllable radiation exposure of the population. This is now clear, nearly hundred years after the discovery of radioactivity. Remediate and preventive activities have been started with a complex approach using:

- building engineering - pasive and active countermeasures, etc.
- geological sciences - geophysics, hydrogeology, pedology, etc.
- physics - measuring devices and methods, radon transport, etc.
- medicine - epidemiology, cancer research, risk perception, etc. in national radon programs based on local activities.

Despite of long experience in radon problems all these approaches need further development, e.g.:

- better indications for site specific mitigation techniques and cost-benefit optimization for them,
- radon mapping of settlements
- a better prognosis for different climatic regimes from measuring results,
- deeper understanding of lung cancer genesis, smoking interference, risk perception by the population,
- etc.



## THE IMPACT OF ICRP 60 RECOMMENDATIONS ON THE DOSE EQUIVALENT IN LOW-AND-HIGH ENERGY NEUTRON FIELDS

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Recent recommendations of ICRP, published in its publication 60, have introduced a modified relationship between the quality factor and unrestricted linearenergy that reflects higher values of the relative biological effectiveness for intermediate neutrons, with its maximum at about 300 to 400 keV. This has resulted in a higher attention that is recently paid to the consequences of these new recommendations for radiation protection quantities in realistic radiation fields with a high contribution of neutrons. A continuous increase in attention has been given to the radiation environment in the nuclear power plants and fuel cycle (1), and to high energy neutrons as occur in stray radiation fields around high energy accelerators and in altitudes of jet aircrafts and space missions (2).

The objectives of this study was to determine the impact of the increased risk factors for neutrons after ICRP 60 on the operational dose equivalent quantities at a few neutron fields selected with respect to cover the broad variety of neutron spectra:

1. Cadarache calibration assembly, with the average neutron energy around 0.6 MeV, designed to simulate realistic neutron spectra at workplaces. This assembly is basically composed of an almost spherical  $^{238}\text{U}$  convertor irradiated by 14.6 MeV neutrons from an accelerator target, placed at its center, and a scattering chamber consisting of a cylindrical polyethylene duct and a series of additional shieldings.

2. Neutron spectra at exposed workplaces in nuclear power plants.

3. Moderated spectra of  $^{252}\text{Cf}$  fission source.

4. Neutron spectra behind a shielding made of the iron (the average energy 5.89 MeV) and concrete (the average energy 46.51 MeV), respectively.

The shielding was located around a thick copper target bombarded by a secondary beam of 205 GeV/c positively charged particles estimated to consist of about 2/3 protons and 1/3 pions.

5. Cosmic rays induced neutron spectra measured on the top of the Zugspitze (2968m) where there is the average neutron energy around 40 MeV.

Except in Cadarache, the neutron spectra were determined by a multisphere spectrometer consisting of a set polyethylene spheres with the following diameters: 2.5, 3, 3.5, 4, 5, 6, 7, 8, 9, 10, 11, 12 and 15 inches. The fluence of thermal neutrons was measured by a  $^3\text{He}$  spherical proportional counter, 32 mm in diameter. The 9" -sphere with polyethylene replaced by lead is used additionally to improve the resolution in high energy region. The response functions were calculated by the Monte Carlo codes MCNP /3/ (below 10 MeV) and HADRON /4/ (above 10 MeV). For the spectra unfolding, the BON 95 /5/ code was used that is based on the parameterization method substantially described /6/. The low energy neutron spectra above the cadmium threshold are described by superposing the 1/E energy tail and the Maxwellian peak with variable temperature and width, whereas the high energy spectra are unfolded by using a combination of the 1/E low energy tail for the low energy peak of evaporated neutrons and the Maxwellian peak, was applied for the high energy cascades. The temperatures of the thermal and cascade peaks were chosen to be 0.035 eV and 55 MeV, respectively.

A Lil cylindrical scintillator was used in the Cadarache measurements. The iterative unfolding code SAND II /7/ was employed to derive spectral neutron fluences.

From the derived neutron spectra, the mean quality factors and conversion factors  $h'$  (10) after ICRP 21 and ICRP 60, respectively, were calculated. The dose equivalent conversion factors were taken from ICRP 51 and /8/ for the energy region below 20 MeV, and from /9/ for the energy region above 20 MeV. The results show that the operational quantities were affected predominantly in the low energy fields, where the changes are given by a factor of 1,3 for the neutron fields given above. As has been expected, the impact of the new recommendations

depends on the shape of the neutron spectra. Therefore, this factor can be much higher in the fields where the intermediate energy region is dominant, which is the case of moderated and scattered spectra at some places in the nuclear power plant and around containers with the spent fuel elements.

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## THE RADIATION PROTECTION OPTIMISATION IN CONTRAST X-RAY DIAGNOSTIC TECHNIQUES

### Abstract

In the class of artificial sources, X-ray diagnostic techniques irradiate global population with more than 90 % share in total dose. At the same time this is the only area with high possibilities in collective dose reduction without important investments. Exposure of the medical team is mainly related to **unnecessary** irradiation. Eliminating this unnecessary irradiation quality of diagnostic information remains undisturbed.

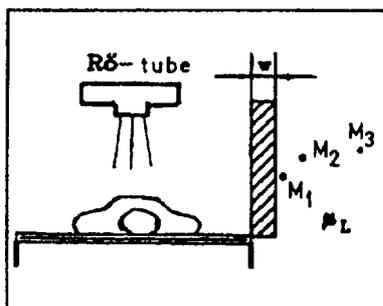
From the radiation protection point of view the most critical X-ray diagnostic method is angiography.

This paper presents the radiation protection optimisation calculation of the protective lead thickness using the Cost - Benefit analysis technique.

The obtained numerical results are based on calculated collective dose, the estimated prices of the lead and lead glass thickness and the adopted price for monetary value of the collective dose unit  $\alpha$ .

### 1. The calculation procedure

The positions of the medical team members shown on fig. 1, are the same, as in the case of the calculation of the medical team irradiation [1].



#### calculation conditions:

$$H_{k,i}(w) < H_L$$

$H_L$  - annual dose equivalent limit for occupationally exposed persons

$H_{k,i}(w)$  - integral dose equivalent for  $k$  - th procedure and

$i$  - th person

Fig. 1. The protection from scattering radiation: added lead slab

The final collective dose equivalent  $S_{E,k}$  from scopy and graphy irradiations and for the life span of equipment of  $\tau$  years, as in the case of the irradiation assessment [1], will be [7]:

$$S_E = \tau \sum_{k=1}^m N_k B_k(\mu w) \left( t_k \sum_{i=1}^n \dot{H}_{0,k,i} + n_{g,k} \sum_{i=1}^n H_{g,k,i} \right) e^{-\mu w} \quad (1)$$

Since the individual doses are far below the limit, health detriment  $Y$  can be expressed simply:

$$Y = \alpha S_E \quad (\alpha = 20\,000 \text{ USD/manSv}) \quad (2)$$

The price of the rectangular shield with area  $ab$  and thickness  $w$  consists of fixed and variable part:

$$X(w) = X_0 + X_v V = X_0 + abwX_v \quad (3)$$

Assuming  $B(\mu w) \approx \text{const}$  and:

$$\alpha \tau \sum_{k=1}^m N_k \left( t_k \sum_{i=1}^n \dot{H}_{0,k,i} + n_{g,k} \sum_{i=1}^n H_{g,k,i} \right) = A \quad (4)$$

the general optimisation condition:

$$\frac{dX(w)}{dw} = - \frac{dY(w)}{dw} \quad (5)$$

becomes finally:

$$w = \frac{1}{\mu} \ln \frac{\mu BA}{abX_v} \quad (6)$$

## 2. Numerical calculations

Assuming:

$$ab = 1 \text{ m}^2,$$

$$X_v = 400 \text{ USD for lead slab of } 1 \text{ m} \times 1 \text{ m} \times 1 \text{ cm},$$

$$E_{\text{eff}} = 60 \text{ keV},$$

$$\mu = 40 \text{ cm}^{-1},$$

$$B = 2.4,$$

$$mN_k = N = 500 \text{ (total number of procedures annually)}$$

$$\tau = 20 \text{ years},$$

and using the originally created analytical approach and computer program (ANGIO80) for calculation of the scattered radiation, the total collective dose of medical team for all procedures and life span of the equipment  $\tau$  is: **1.6 man Sv**.

Finally, using Eq. 6 the optimal thickness becomes:

$w = 2.2$  mm of lead

$m = 25.5$  kg,

attenuation factor  $e^{aw} = 6\ 630$

So unexpectedly thick layer (and high attenuation factor) is caused by the extremely low expenses assumed in comparison with value  $\alpha$ .

Adding new expenses (lead glass window, holes for hands, slider, amortisation...) the price of the lead slab increases and the thickness decreases. For the added expenses of 2 000 USD, the new optimal thickness becomes:

$w = 1.6$  mm ( $e^{aw} = 600$ ,  $m = 19$  kg)

THIS DECREMENT OF THE PROTECTION LEVEL DUE TO INCREMENT OF THE EXPENSES DESCRIBES THE ESSENCE OF THE OPTIMISATION

### 3. The new proposition for protection from scattered radiation: the sliding screen

In the publication *Manual on Radiation Protection in Hospitals and General Practice, Vol. 3, X - Ray Diagnosis, WHO, Geneva, 1975, [2]*, the authors suggest the hanged screen for protection of scattered radiation presented on figure 2.

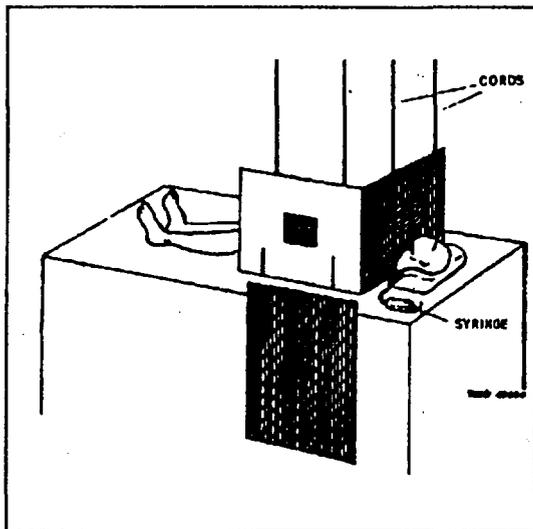


Fig.2. The hanged screen for protection of scattered radiation

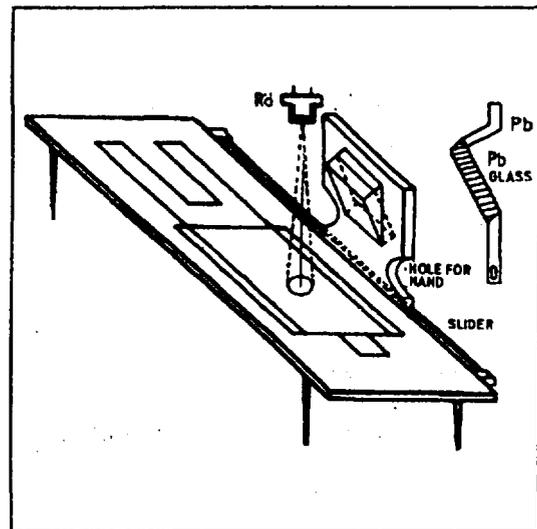


Fig.3. A new proposition: the sliding screen with the aslant mounted lead window and holes for hands

In our opinion, this might not be the best solution, especially from the patient point of view. Such designed protecting device is uncomfortable. We propose the construction shown on

figure 3. Taking in to account the attenuation factor of this lead slab, the question: " *is the lead apron really necessary ?*" seems to be reasonable.

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## THE SIMPLE ANALYTICAL METHOD FOR SCATTERED RADIATION CALCULATION IN X - RAY DIAGNOSTIC TECHNIQUES

### Abstract

In realization of radiation protection measures for medical staff present during diagnostic procedures, the necessary condition is knowledge of the space - energy distributions of the scattered radiation from the patient.

In this paper, the simple calculation procedure for the scattered radiation field of the actual diagnostic energies is presented.

Starting from the single Compton scattering model and using the justified transformations the final equations in elementary form are derived.

For numerical calculations the computer code ANGIO was created.

The calculated results were confirmed by detailed dosimetric measurements of the scattered radiation field around patient (the water phantom) in SSDL in the Institute of nuclear sciences "Vinča", Belgrade. These results are good base for assessment of irradiation.

The main irradiation source for the physician and the other members of the medical team is the back scattered radiation from patient - albedo.

### 1. The analytical approach

The scattering geometry is shown on fig. 1. The target is the head of the physician. On fig. 2. the active part of the scattering volume is shown. The elementary volume on the depth  $z$  can be calculated by the equations 1.

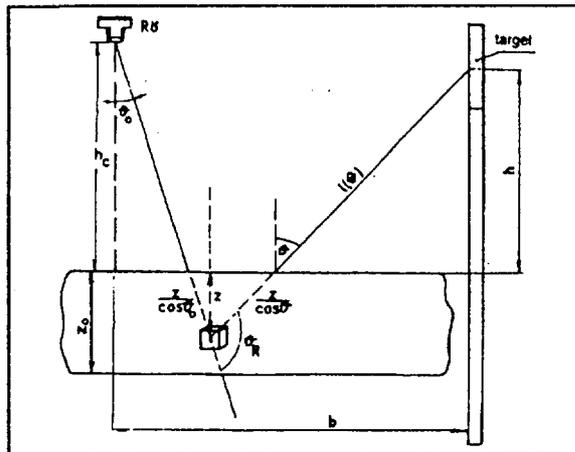


Figure 1. Scattering geometry

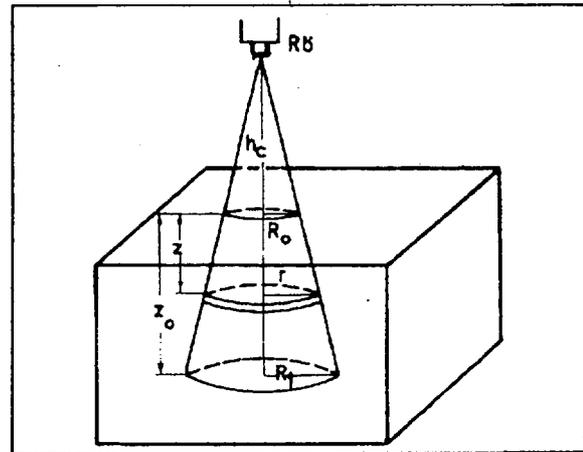


Figure 2. Active part of scatterer

$$\begin{aligned}
 dV &= \pi r^2 dz \\
 dz &= \frac{h_c}{R_0} dr \\
 dV &= \frac{\pi h_c}{R_0} r^2 dr
 \end{aligned} \tag{1}$$

Calculating the probabilities for penetration of the initial photon without interactions to depth  $z$  through scatterer, for experiencing the Compton scattering at the depth  $z$  into unit space angle defined with  $\theta$  and  $\phi$  and distance  $l(\theta)$ , and for scattered photon to hit the target without any further interaction, one can obtain the starting equation for space-angular distribution of the scattered radiation:

$$dI_R(\Omega) = I_0 B n_e \frac{d_e \sigma(\theta_R)}{d\Omega} e^{-z(\frac{\mu_1}{\cos\theta_0} + \frac{\mu_2}{\cos\theta})} \frac{dV}{l^2(\theta)} \tag{2}$$

where are  $dV$  - elemental volume around the scattering point,  $l(\theta)$  - scattering point target distance.

Using eq. 1. and substituting variable  $r$  with  $z$ , one can obtain:

$$I_R(\Omega) = C_1 \int_0^{z_0} \frac{(z+h_c)^2}{l^2(\theta)} \frac{d_e \sigma(\theta_R)}{d\Omega} e^{-z(\frac{\mu_1}{\cos\theta_0} + \frac{\mu_2}{\cos\theta})} dz \tag{3}$$

where  $C_1 = \frac{I_0 B n_e \pi R_0}{h_c^2}$ , and  $z_0$  - maximum penetration depth in tissue.

After several justified approximations for actual energy ranges in tissues we shall finally obtain the scattered intensity (the energy flux density) which hits the target through the unit space angle (defined with  $\varphi_R$  and  $\theta_R$ ):

$$I_R(\Omega) = \frac{C_1 d_e \sigma(\theta_R)}{b^2 d\Omega} \sin^2 \theta \cdot I_1(\theta) \quad (4)$$

where:

$$I_1(\theta) = \frac{h_c^2}{\mu(1+\sec\theta)} + \frac{2h_c}{\mu^2(1+\sec\theta)^2} + \frac{2}{\mu^3(1+\sec\theta)^3} - e^{-\mu z_0(1+\sec\theta)} \left( \frac{(z_0+h_c)^2}{\mu(1+\sec\theta)} + \frac{2(z_0+h_c)}{\mu^2(1+\sec\theta)^2} + \frac{2}{\mu^3(1+\sec\theta)^3} \right) \quad (5)$$

The considerable simplification of the calculating procedure one can perform substituting the truncated cone by the cylinder. After some transformations, we finally obtain:

$$I_R(\theta) = \frac{C_2 d_e \sigma(\theta)}{l^2(\theta) d\Omega} \cdot \frac{1 - e^{-\mu z_0(1+\sec\theta)}}{\mu(1+\sec\theta)} \quad (6)$$

Since the interaction probabilities strongly depend on the energy of the initial photon, the multigroup technique in treatment the spectrum was used. For numerical calculation the computer code ANGIO is created.

Numerical results are tested by the detailed dosimetric measurements for two original spectrums:

1. U=60 kV I=10 mA, filter 4 mm Al + 0,3 mm Cu, HVL=0,18 mm Cu
2. U=80 kV I=10 mA, filter 4 mm Al + 0,5 mm Cu, HVL=0,35 mm Cu

The calculated and measured results for both spectra (60 kV and 80 kV) are presented on fig. 3. Each point in the space measurement net contains 4 results: the calculated and measured values for both of voltages. From fig. 3. one can establish very good agreement between calculated and measured values:

- inside 10 % for 60 kV, and
- inside 5 % for 80 kV.

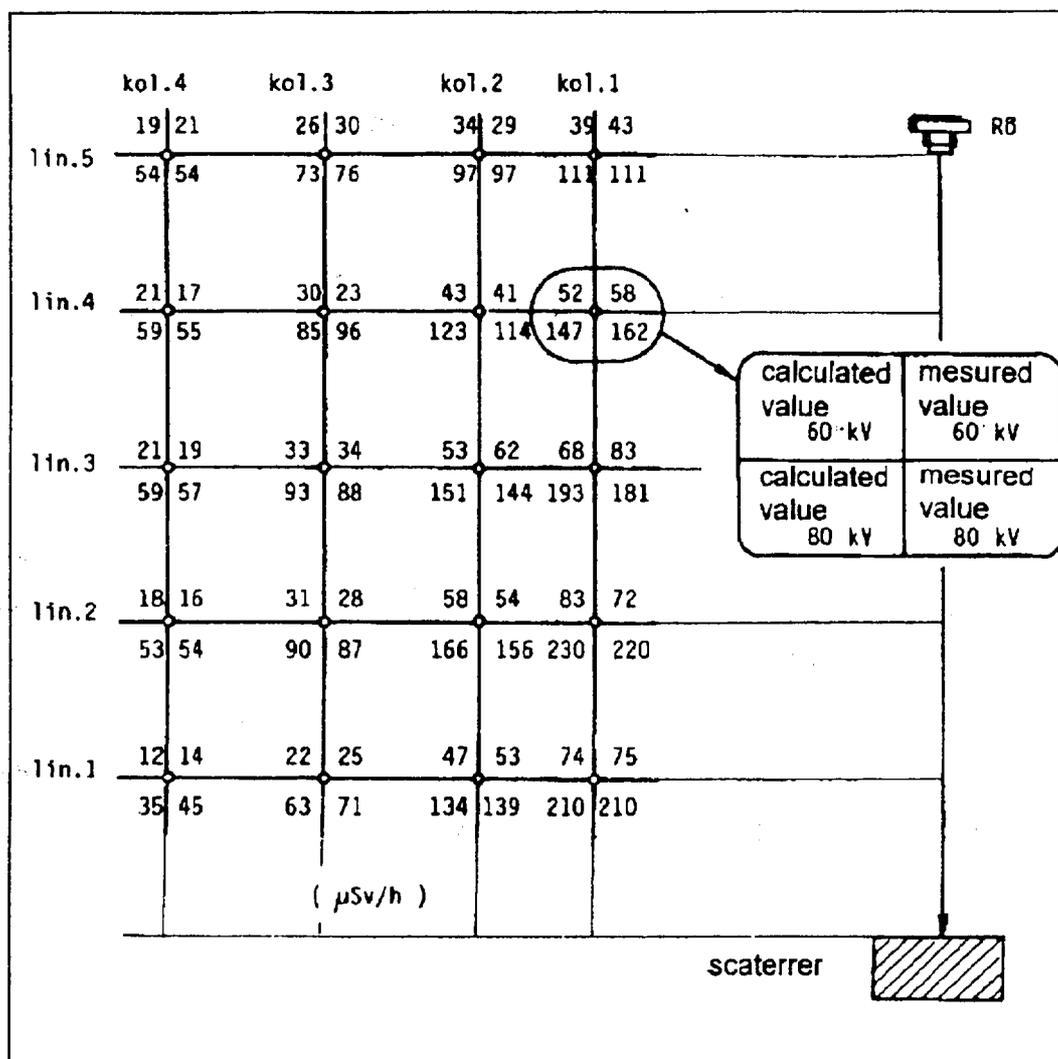


Figure 3. Measurement net with results

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## THE EVALUATION OF THE IRRADIATION OF MEDICAL TEAM IN CRITICAL X-RAY DIAGNOSTIC TECHNIQUES

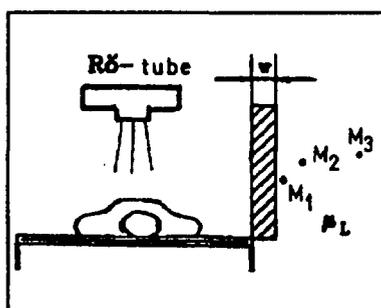
### Abstract

A good realized assessment of the irradiation for any exposed group of population serves as the base for the radiation protection measures (emergency radiation preparedness, radiation protection optimisation etc.). This is especially important, by the radiation protection point of view, in contrast X - ray diagnostic techniques - angiographies.

This paper presents the way for the realization of the medical team irradiation assessment, based on originally derived simple equations for the scattered radiation field around patient.

### 1. The collective dose calculation

A new approach for the radiation protection of the medical team is presented on figure 1. The lead slab with thickness  $w$  and attenuation coefficient  $\mu_L$  shields from scattered radiation. The positions of physician, assistant and technician are marked by  $M_1$ ,  $M_2$  and  $M_3$  respectively.



**condition:**

$$H_{k,i}(w) < H_L$$

$H_L$  - annual dose equivalent limit for occupationally exposed persons

$H_{k,i}(w)$  - integral dose equivalent for  $k$  - th procedure and

$i$  - th person

Fig. 1. The protection from scattering radiation: added lead slab

Adding lead shield with thickness  $w$ , the equivalent dose rate in position  $M_1$  will be:

$$\dot{H}_{k,i}(w) = \dot{H}_{0,k,i} B(\mu w) e^{-\mu w} \quad (1)$$

$\dot{H}_{0,k,i}$  - dose equivalent rate in air for k-th procedure and i-th person

$B(\mu w)$  - build up factor.

The collective dose equivalent  $S_{E,k}$  during k-th procedure consists of doses received from scopy and graphy irradiations:

$$S_{E,k} = B_k(\mu w) \left( t_k \sum_{i=1}^n \dot{H}_{0,k,i} + n_{g,k} \sum_{i=1}^n H_{g,k,i} \right) e^{-\mu w} \quad (2)$$

$B_k$  - build up factor for k-th procedure

$H_{g,k,i}$  - integral dose equivalent for one graphy for i-th person and k-th procedure

$n_{g,k}$  number of graphies in k-th procedure.

If  $N_k$  is number of procedures of k-th type annually, then for m types of procedures, the annual collective equivalent dose  $S_{E,g}$  will be:

$$S_{E,g} = \sum_{k=1}^m N_k S_{E,k} \quad (3)$$

and for the life span of equipment of  $\tau$  years:

$$S_E = \tau \sum_{k=1}^m N_k B_k(\mu w) \left( t_k \sum_{i=1}^n \dot{H}_{0,k,i} + n_{g,k} \sum_{i=1}^n H_{g,k,i} \right) e^{-\mu w} \quad (4)$$

## 2. Numerical calculations

Assuming:

$$ab = 1 \text{ m}^2,$$

$X_v = 400$  USD for lead slab of 1 m x 1 m x 1 cm,

$$E_{\text{eff}} = 60 \text{ keV},$$

$$\mu = 40 \text{ cm}^{-1},$$

$$B = 2.4,$$

$mN_k = N = 500$  (total number of procedures annually)

$$\tau = 20 \text{ years}$$

and using the originally created analytical approach and computer program (ANGIO80) for calculation of the scattered radiation, the equivalent dose rates for physician, assistant and technician are 200, 150 and 50  $\mu\text{Sv/h}$  respectively.

The contribution to the collective dose without added protecting slab, for scopy, will be:

$$t_k \sum_{i=1}^n \dot{H}_{0,k,i} = 0.2h(200+150+50) \mu\text{Sv}/h = 80 \text{ man}\mu\text{Sv} \quad (5)$$

Dividing the measured patient skin dose rate ( $\mu\text{Sv}/h$ ) by number of mA and by 3 600 (seconds per hour) one can obtain dose per mA and per second. Taking the same scattering coefficient as in the case of scopy and assuming 36 spent films per one procedure the contribution to the collective dose from graphy becomes: **80 man  $\mu\text{Sv}$ .**

The annual collective dose per one procedure becomes: **160 man  $\mu\text{Sv}$ .**

For 500 procedures annually and 20 years of life span of equipment the total collective dose for medical team will be: **1.6 man Sv**

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## RADIATION PROTECTION AND DOSIMETRY PROBLEMS AROUND MEDIUM ENERGY ACCELERATORS

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### Abstract

In the Institute of nuclear sciences "VINČA", the Accelerator Installation "TESLA", which is an ion accelerator facility consisting of an isochronous cyclotron "VINCY", a heavy ion source, a D<sup>-</sup> / H<sup>-</sup> ion source, three low energy and five high energy experimental channels is now under construction.

The Tesla Accelerator Installation should be the principal facility for basic and applied research in physics, chemistry, biology, and material science, as well as for production of radioisotopes, medical diagnostics and therapy with radioisotopes and accelerated particle beams.

Some problems in defining radiation protection and safety programme, particularly problems in construction appropriate shielding barriers at the Accelerator Installation "TESLA" are discussed in this paper.

**Key words:** accelerator facility, radiation protection, shielding

### Introduction

As it has been said, the VINCY cyclotron is an isochronous cyclotron. In creating its concept the main aim was to obtain a multipurpose machine, i.e. to enable the acceleration of ions in a wide range of specific charges. The first requirement was to obtain heavy ions with energies well above the Coulomb barrier, i.e. well above 5 MeV per nucleon, and make the machine a good instrument for research in nuclear physics. The second requirement was to obtain sufficiently high intensities of protons and deuterons with energies above  $\approx 60$  MeV, and make the machine a sufficiently good instrument for applications in medicine. High-energy heavy-ions are planned in use for in-situ analysis of the elementary process in the materials exposed, including research on microdosimetry and track structure and energy deposition of accelerated particles.

The VINCY cyclotron will be able to deliver  $\approx 1 \mu\text{A}$  of 36 MeV per nucleon O<sup>8+</sup> ions,  $\approx 100 \text{ nA}$  of 23 MeV per nucleon Ar<sup>16+</sup> ions and  $\approx 700 \text{ nA}$  of 7 MeV per nucleon Xe<sup>28+</sup> ions. It will be able to deliver also heavy ions of lower energies - above  $\approx 3$  MeV per nucleon. This machine will be able to give  $\approx 20 \mu\text{A}$  of 73 MeV deuterons, and  $\approx 2 \mu\text{A}$  of 66 MeV protons. It will be able to give also deuterons of lower energies - above  $\approx 43$  MeV, and protons in the energy region of 22 - 36 MeV. The principal schema of the TESLA Accelerator installation is shown in Figure 1.

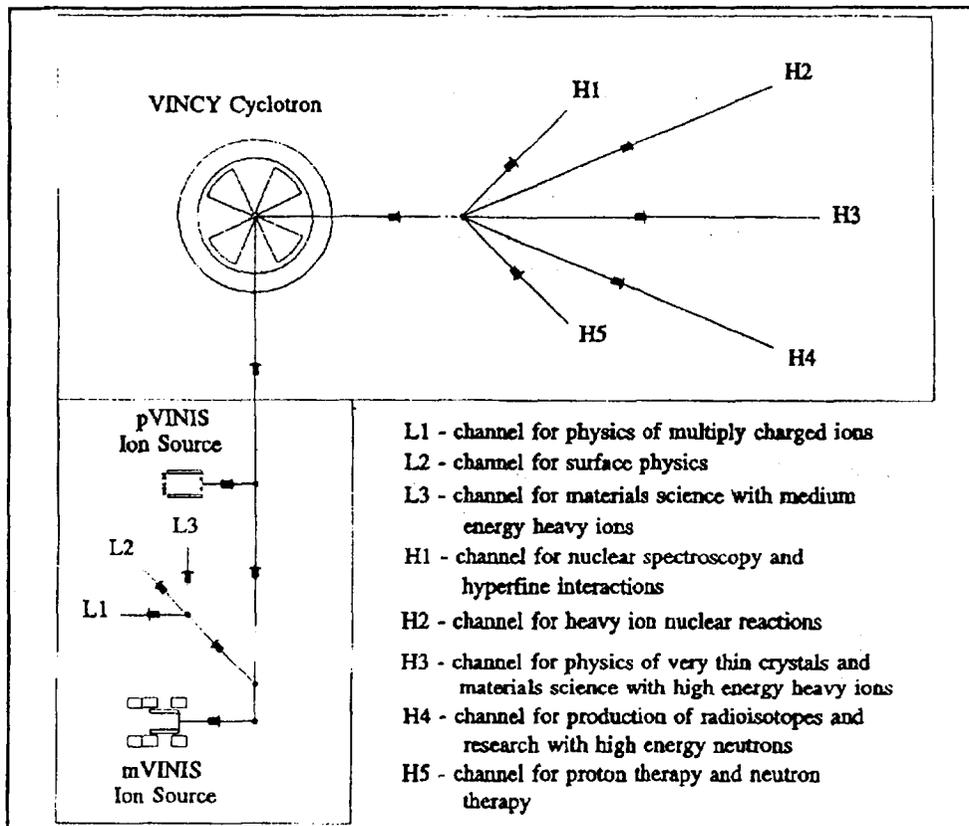


Figure 1. The principal scheme of the TESLA Accelerator Installation (TAI 93)

### Radiation environments of particle accelerators

Two distinct and separate radiation fields are associated with particle accelerator, and both are of practical concern to health physicist. The first may be described as "prompt", and is directly associated with operation of accelerator. All components of this prompt radiation field disappear almost immediately upon accelerator turn off. The second radiation field described as "remanent" since it remains after accelerator operation has ceased; it is due to radioactivity induced in the accelerator structure (PAT 73).

The prompt radiation field is produced either by the atomic or nuclear interaction of particles during acceleration, or in the utilization of accelerated particles. Inefficiencies in acceleration process lead to particle losses during the acceleration cycle. If these particles have sufficient energy they may induce nuclear interaction in the accelerator structure, generating a short-lived radiation field the detailed composition of which is determined by the energy and type of the accelerated particles and the material in which they interact. Beam losses during the acceleration cycle may place severe limitations on beam intensity or may necessitate substantial radiation shielding.

Although beam losses may be important, they represent only a small fraction (typically a few

percent) of the useful accelerator beam power. In use the accelerator beam interacts with an experimental target, irradiated specimen, or patient, and it is these interactions that are largely responsible for the general character of the radiation field (PAT 73, TRS 88).

Full understanding of the prompt radiation field generated requires knowledge of the primary interaction in the target material and the subsequent progression of the interaction products through the structure and surrounding experimental material and shielding.

Health physics problems at heavy-ion accelerators, with exception of accidental exposure close to or directly in the accelerator beam, are largely due to their neutron production. Thus an understanding of neutron production by charged particles is an essential weapon for the armoury of the health physicist, who must design shielding for such accelerators, estimate their production of radioactivity, and measure the radiation field they produce.

### **Shielding criteria for medium energy particle accelerators**

All existing medium and high energy accelerators require some shielding for two reasons:

- to limit the biological effects on occupationally exposed persons and population exposure to acceptably values, according to basic radiation protection principle (ALARA - As Low As Reasonable Achievable (ISR 91, BSS 94)),
- to reduce experimental background conditions to tolerant values.

The design of a practical shielding arrangement for medium and high energy accelerator is in general a rather complex task for which no simple hand-book formulae are adequate. Nevertheless, it is possible by using a series of methods to make reasonably precise design in many instances and to allow for an adequate range of design in many other cases (KOM 86).

At its best the calculation for the shielding of a medium, as well as high energy accelerators involves, many simplifying assumption to make the problem tractable. Nevertheless the magnitude of the cost at stake and the engineering problems of support and handling forbid the casual use of large margins of safety, and require an attempt at precision compatible with the knowledge of the basic factors of production and propagation of radiation.

It is a fact familiar to all who have considered the shielding of a heavy particle accelerators that the radiation component that dominate in the prompt radiation field and the shielding problem, on the accelerator with performances like the TESLA Accelerator Installation have, is neutron radiation.

The beam losses interacting with components of accelerator structure, as well as beam of accelerated particles interacting with experimental targets, irradiated specimen or patient will give rise to high neutron fluxes, for which shielding must be provided, and also lead to component damage and induced radioactivity, with the attendant risk of increased radiation exposure of maintenance personnel.

Experimental data on neutron yields and spectra from stopping protons are available at proton energies above 100 MeV; however, there is much less data for protons and for deuterons in the energy range between 50 and 100 MeV, where inelastic cross sections change rapidly with energy (BRO 83, FAS 76).

### Conclusion

The goal of all efficient accelerator shielding design is to attenuate the high radiation intensities produced by the accelerator and its associated equipment to levels that are acceptable outside the shielding, at minimum cost and without compromising the utility of the particle accelerator for its designed purposes. The first and the most important and the most complicated step is determination of radiation sources, i.e. the space, time, particle and energy distribution of radiation fields (NCR 77). It is, therefore the most complicated step in solving radiation protection and shielding problems also in TESLA Accelerator Installation, especially due to the substantial lack of information about interaction cross section for the most important accelerated particles - protons and deuterons, in the energy range of interest (BRO 83).

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## SOME SOLID STATE PROPERTIES OF LiF:Mg,Cu,P TL-MATERIAL

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### INTRODUCTION

LiF:Mg,Cu,P is a very interesting thermoluminophor for individual and environmental monitoring because it combines a high sensitivity to radiation with low energy dependence of the TL response for exposures to photon radiation. The thermoluminophor LiF:Mg,Cu,P is commercially available as powder and in form of sintered pellets or fixed onto a polyimide (Kapton<sup>R</sup>) tape. The dosimetric properties of this thermoluminophor has been reported by various authors [see e.g. ref. 1-5]. Besides the advantages in view of high sensitivity, low energy dependence and wide linearity of dose characteristics a general problem of LiF:Mg,Cu,P TL material is that the TL sensitivity can decrease under heat treatment at temperatures higher than 260 °C and that short-term annealing below 240 °C are not satisfactory for total removing the residual TL-signal.

These effects are investigated intensively with the aiming at finding procedures which permit a re-use of the dosimeters without a loss of sensitivity. Only some papers give an explanation about this remarkable behaviour of this TL-material on the basis of a LiF-matrix [6,8].

This paper describes some investigations of solid state characteristics of a LiF:Mg,Cu,P thermoluminophor.

### MATERIAL AND METHODS

The investigations were carried out with LiF:Mg,Cu,P-TL-material prepared by the chemical institute of the Moscow State University in form of powder and sintered pellets. Following methods were use:

- Studies of the chemical composition was carried out by x-ray fluorescence analysis with „SPECTRO-X-LAB“-equipment (Spectro x-Ray Instruments) with Rh-anode, B<sub>4</sub>C-polarizator, LN<sub>2</sub>-cooled 30 mm<sup>2</sup> Si(Li)-Detector with Be-window (energy resolution 155 keV for Mn-k<sub>α</sub>-radiation). The software of the equipment permits a qualitative and quantitative determination of elements with atomic numbers > 10.
- Crystal structure investigations were taken by x-ray-diffractometry with a SIEMENS-diffractometer D 5000 using Cu-k<sub>α</sub>-radiation. The integrated software permits to analyze the crystalline phases using the data of the measured material by comparison with standard spectra of various pure substances.

### RESULTS OF MEASUREMENTS

#### Quantitative analysis of the chemical composition

The determined contents of the dopants Mg, Cu and P and of other elements with Z > 10 are summarised in tab.1. The contents of Mg and Cu are typical concentrations of impurities with dopant character. The P-content is very high and has the character of a second matrix component that can form various lithium phosphate compounds. Other elements with potential

dopant concentrations are Si, Al, and Na, further Fe and Mn with lower ppm-values. The content of all other elements are lower than 3 ppm.

### **Crystal structure determination**

Figures 1 and 2 show spectra of x-ray diffraction measurements. Naturally, the diffraction lines of the LiF-basic material dominate in the diffraction spectrum. However, also the lines of  $\text{Li}_3\text{PO}_4$  and  $\text{Li}_4\text{P}_2\text{O}_7$  appear significantly. Two diffraction lines were detected in the spectrum of LiF:Mg,Cu,P that do not permit an identification of a definite compound using a standard spectrum. These lines do not fluctuate if the material was heated up to 400 °C and had lost its high TL-sensitivity.

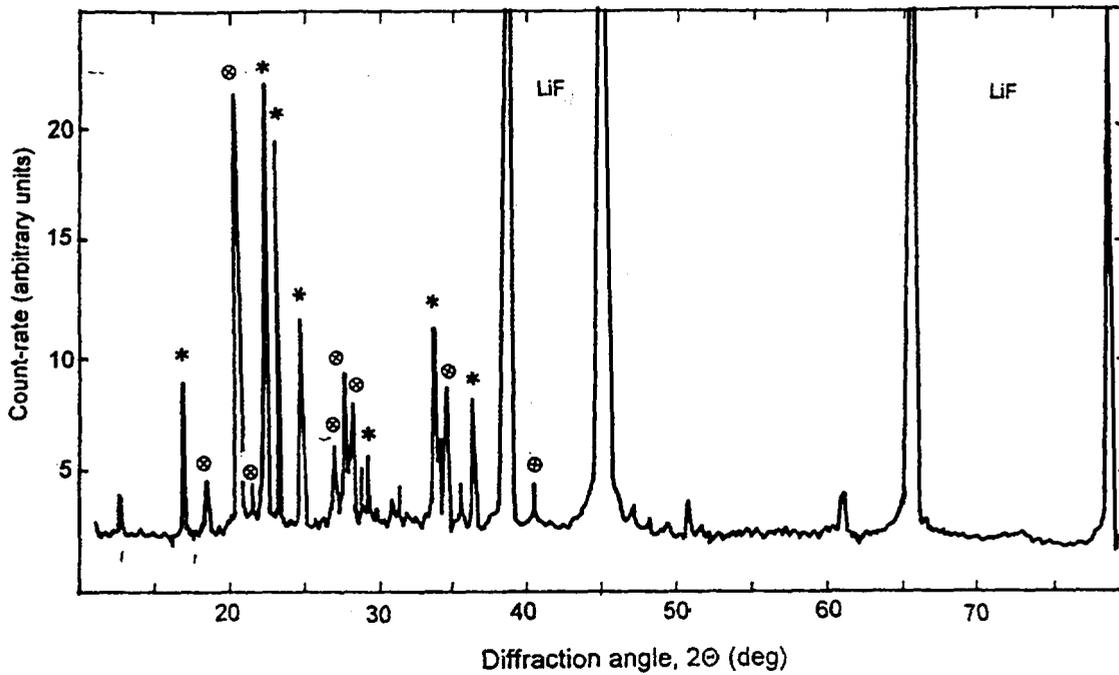
### **DISCUSSION OF THE RESULTS AND CONCLUSIONS**

The results of determination of the chemical composition and the crystal structure show that in the thermoluminophor LiF:Mg,Cu,P, besides the basic material LiF also  $\text{Li}_3\text{PO}_4$ - and  $\text{Li}_4\text{P}_2\text{O}_7$ -crystal regions exist. The occurrence of the two lithium phosphate phases follow from the high ammonium phosphate content in the mixture for the thermoluminophor production. The formation of the various lithium phosphates depends from state of dehydration of phosphoric acids, created by thermal decomposition of  $\text{NH}_4\text{H}_2\text{PO}_4$ , before their reactions with LiF start. Therefore the content of these compounds can differ if thermoluminophors are prepared under various conditions (e.g. temperature, time and rate of heating, sample size, mixture status). This assumption can explain that SUN et al.<sup>[8]</sup> identified only lines of  $\text{Li}_4\text{P}_2\text{O}_7$ . The two not identified lines in the X-ray diffraction spectrum are with high probability also a crystal phase formed by Li, F and a phosphate, because the low content of other impurities render hardly the formation of a own crystal structure. SUN et al.<sup>[8]</sup> observed in another LiF:Mg,Cu,P-material also lines of an unidentified „X material“. In contrast to the proposal of these authors it seems that the unknown compound in our material is not the reason of its high TL sensitivity, because this phase also exists if the strong TL-effect was destroyed by heating of the sample up to 400 °C.

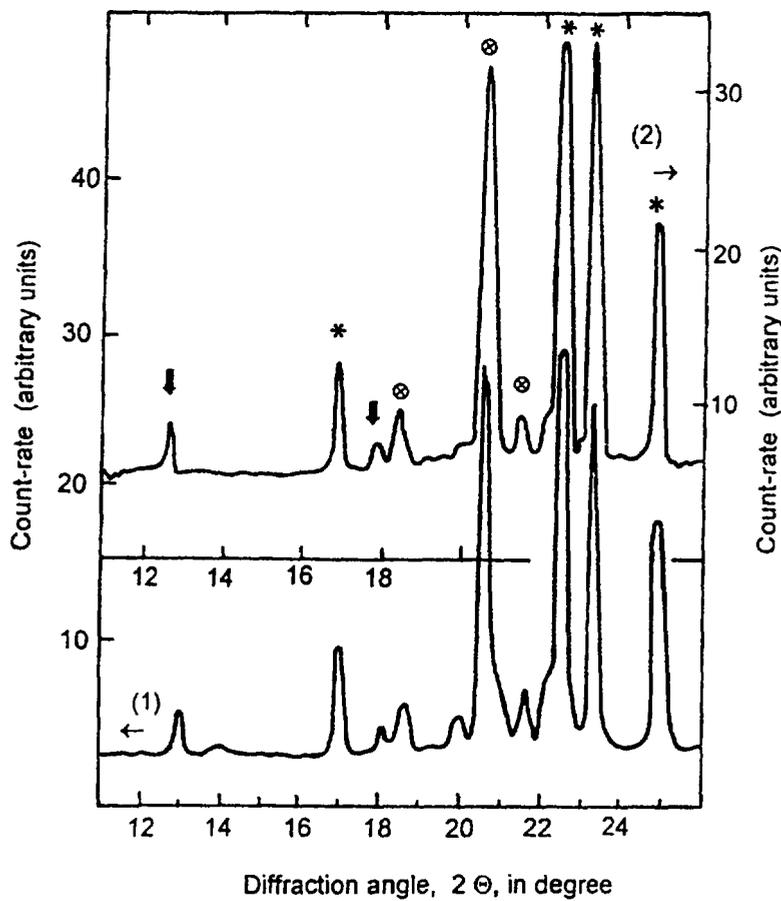
The thermoluminophor LiF:Mg,Cu,P is in contrast to LiF:Mg,Ti a material consisting of different chemical compounds and crystal structures. It seems that a strong TL-effect appears only in the case of a special preparation procedure that permits to obtain a optimal mixture of the different structures including an optimal distribution of the dopants Cu and Mg. It is complicate to achieve the optimal state. Therefore the properties of various production batches were not exactly equal. The thermal sensitivity of this structure caused the possibility of the TL-efficiency decreasing in the case of repeated use of the dosimeters. The possibility to change the glow peak shapes by annealing processes of LiF:Mg,Cu,P-pellets permits the suggestion that a thermal sensitive equilibrium state of Li-phosphate structures exists in a LiF matrix. The maintenance of the needed equilibrium of special structures in the material depends on the preparation procedure, on the reading and annealing methods. Typically for such an equilibrium is its poor thermal stability.

The following detector properties can therefore explained:

- Batch dependence regarding TL-sensitivity and TL-response stability,
- strong decrease of TL-response after heating higher than certain temperature



**Fig.1** Spectrum of x-ray diffraction of LiF:Mg,Cu,P TL material  
 \* and ⊗ indicate the diffraction spectrum lines of  $\text{Li}_3\text{PO}_4$  and  $\text{Li}_4\text{P}_2\text{O}_7$  respectively which appear besides the lines of LiF



**Fig.2** Spectrum of x-ray diffraction of LiF:Mg,Cu,P TL material, region of Li-phosphate lines. (1) Virgin material, (2) material after an annealing procedure at 400 °C.  
 \* and ⊗ indicate the diffraction spectrum lines of  $\text{Li}_3\text{PO}_4$  and  $\text{Li}_4\text{P}_2\text{O}_7$  respectively. The arrows mark the unknown component lines

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Tab.1. Dopants and impurities in LiF:Mg,Cu,P-TL-material

| Status     | Element                 | Concentration in ppm |
|------------|-------------------------|----------------------|
| Dopants    | P                       | 63500 ± 800          |
|            | Mg                      | 1500 ± 100           |
|            | Cu                      | 850 ± 5              |
| Impurities | Si                      | 6580 ± 300           |
|            | Al                      | 1860 ± 50            |
|            | Na                      | 1500 ± 970           |
|            | Fe                      | 88 ± 5               |
|            | Mn                      | 77 ± 4               |
|            | Σ other elements Z > 10 | 3                    |



# $^{90}\text{Sr}$ LEVEL IN FRESH AND POWDER MILK IN SOME AREAS OF ALBANIA CAUSED BY THE CHERNOBYL ACCIDENT

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## SUMMARY

Chernobyl accident caused a rather high level of territory contamination, especially in the east and north-east areas of Albania. That's why for the radioprotection scope of population in the INP, during and after the accident a great number of environmental samples are measured. An important type of samples were those of the fresh and powder milk. Except the  $^{131}\text{I}$  and  $^{137}\text{Cs}$  radioactivity concentration measurements,  $^{90}\text{Sr}$  level in milk determination was performed.

The tracing method by stable strontium was used for the chemical separation ratio determination.

The  $^{90}\text{Sr}$  activity concentration in milk was evaluated by  $^{90}\text{Y}$  in equilibrium activity measurement. The dynamics of the  $^{90}\text{Sr}$  in milk for a period of some months, just after the accident and some daily measurements carried-out during the "hot" period are given in this paper.

## INTRODUCTION

Parallelly with the widening of the nuclear power and radioactive isotopes use, the environmental radioactive contamination measurement are development for radiation protection scope.

As the result of the food chain grass-cow-milk-human the environmental radioactive materials can be transferred in human body. The presence of  $^{90}\text{Sr}$  in biosphere is a long-term risk, due to its chemical similarity to calcium and its long biological half-life [1]. Since 1978, some environmental measurement methods for beta and beta-gamma low-level measurement are carried-out. Global beta counting with proportional gas detectors, liquid scintillation counter and low-level gamma spectrometry are some of them [2]. Concerning the  $^{90}\text{Sr}$ , taking into account the level and the fact both  $^{90}\text{Sr}$  and its daughter  $^{90}\text{Y}$  are pure beta

emitters the general measurement procedure are based on chemical separation and low-level beta counting.

## METHODS

The following milk samples destruction method is used:

- 1- The transference of 1000 ml collected sample in a beaker, adding 4 ml acetic acid ( $\text{CH}_3\text{COOH}$ ) and mixing thoroughly.
- 2- The transference of the acidic milk in two 500 ml porcelain bowls and evaporation at low temperature to avoid the boiling until to dry up.
- 3- The ashing of all dried material in a muffle furnace, raising slowly the temperature up to  $300^\circ\text{C}$  (3 hours) and after that up to  $600^\circ\text{C}$  (15 hours).
- 4- The dissolution of the ash in 45 ml  $\text{HNO}_3$  (65%), sending to 100 ml volume by distilled water and covering with a watch glass.[3].

The separation method used was as follows:

The separation of alkaline-earth elements as phosphate, the nitric acid fuming method to move the Ca ions, and the dregs method to move the Ba ions as chromates is applied. [4].

The tracing by nonradioactive strontium salt of the strontium nitrate waterfree  $\text{Sr}(\text{NO}_3)_2$  was used to determine the chemical separation ratio.

The chemical separation ratio of calcium ( $\text{Ca}^{2+}$ ) ions was determined by the X-ray fluorescence measurement method. It was obtained a higher than 0.99% value.

The overall chemical yield of  $^{90}\text{Sr}$  was 70-80%, while that of  $^{90}\text{Y}$  was 96.8-99.7%. The  $^{90}\text{Sr}$  activity was determined by the  $^{90}\text{Y}$  activity in equilibrium measurements, which sources were produced in oxalate form. The measurements were performed mainly in a low-level beta counting system with proportional gas detector and anticoincidence protection. A LSC system, using the Čerenkov radiation measurements was used, too. The lower limit of detection of the first system was 12 mBq/l, while the registration effectivity was 0.41.

The uncertainty of  $^{90}\text{Sr}$  activity concentration in milk was estimated about 30%, at 68% confidence level.

## RESULTS AND CONCLUSIONS

The  $^{90}\text{Sr}$  activity concentration levels of three sampling areas, dislocated in Tirana (centre of Albania), in Shkodra (north of the country) and Tropoja (north-east) are in Tab.1 represented.

Tab.1

<sup>90</sup>Sr activity concentration on milk

| Place              | 1986 |      |     |     |     |     | 1987 |     |     |     |     |     |      |     |     |      |     |     |
|--------------------|------|------|-----|-----|-----|-----|------|-----|-----|-----|-----|-----|------|-----|-----|------|-----|-----|
|                    | May  | Aug  | Sep | Oct | Nov | Dec | Jan  | Feb | Mar | Apr | May | Jun | Jul  | Aug | Sep | Oct  | Nov | Dec |
| Tirana<br>(Bq/l)   | 4.6  | -    | .16 | .40 | .21 | .19 | .22  | .20 | .21 | .25 | .14 | .05 | .045 | .04 | .13 | .05  | .07 | .06 |
| Shkodra<br>(Bq/kg) | 23   | 1.87 | 1.4 | -   | -   | -   | -    | -   | 1.5 | -   | 1.0 | 1.3 | .80  | .88 | .70 | 1.30 | -   | .45 |

\* long term average value on fresh milk before Chernobyl was 60 mBq/l

\*\* " " " " " on powder milk " " " " 450 mBq/l

Some values of the fresh milk from the "hot" period are given in Tab.2

Tab.2

| Date of the May<br>1986        | 4   | 6    | 8   | 9   | 13 | 14  | 15 |
|--------------------------------|-----|------|-----|-----|----|-----|----|
| Tirana (Bq/l)<br>(cow milk)    | 1.2 | 35.5 | -   | 1.4 | -  | 7.6 | -  |
| Tropoja (Bq/l)<br>(sheep milk) | -   | 55.7 | 7.1 | 3   | 10 | -   | 4  |

The considerable <sup>90</sup>Sr contamination of the fresh milk on May 1986, shows a monthly average of about 80 times greater than the long-term value before the Chernobyl accident.

The absolute maximum obtained around the 6 May was about 1000 times greater than the "normal" value. This is in accordance with the maximum level of the air contamination reached the first days of the May 1986. [2],[5],[6]. At the same time, it can be said that the first maximum of 6 May and the second one, that of 13-14 May is in accordance with the maximum of the contamination of fall-out and that of <sup>131</sup>I in milk. [2],[5],[6]. Another conclusion similar with that of the previous publications is that the north-east part of the country was more contaminated than the inner part, confirming again that the contaminated air streams penetrated in Albania through the east and north-east border of the country.



**ASSESSMENT OF OCCUPATIONAL EXPOSURES TO EXTERNAL  
RADIATION-  
- IAEA RECOMMENDATION 1995**

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**Extended Abstract**

The IAEA recommendation contains the guidance on:

- establishing monitoring programmes,
- the interpretation of results,
- records keeping,
- quality assurance

The objectives for workplace monitoring including the recommended methods are also involved.

The operational quantity defined for individual monitoring is the personal dose equivalent  $H_p(d)$ . For weakly penetrating and strongly penetrating radiations recommended depths are respectively 0.07 and 10 mm  $H_p(0.07)$  and  $H_p(10)$ . In order to design a programme of individual monitoring there is a need to identify the individuals working in controlled areas where individual monitoring is required. For supervised areas the individual monitoring may be only good practice for the purpose of dose records. Where individual monitoring of workers is required, an approved monitoring service, whose quality of services has been approved by an authoritative body, should be selected. The service is responsible for the accuracy and reliability of the service and should be able to evaluate dosimeters within a short time if an overexposure is indicated or expected.

The choice of personal dosimeter depends not only on the type of radiation but also on the method of interpretation to be used:

- photon dosimeters giving information only on the personal dose equivalent  $H_p(10)$  - mostly TL or RPL dosimeters are used
- photon dosimeter of discriminating type giving, in addition to  $H_p(10)$  and  $H_p(0.07)$ , some indication of radiation type and effective energy and detection of electrons - data which must be known for E calculation - mostly film badge is used
- extremity dosimeters giving information on  $H_p(0.07)$  - mostly TL dosimeters are used
- neutron dosimeters giving information on  $H_p(10)$  - track-etch or albedo dosimeters are used.

The monitoring service should have quality assurance testing which is an organization's internal system of procedures and practices which assures the quality of its service. This process may be part of the approval performance testing which is a part of approved procedures carried out by the authoritative organization in regular intervals.

The approved monitoring service should perform the dose records keeping which serve the protection of the workers and these data are the part of the Register of the Professional Exposures which is mostly organized by the authoritative body.



## THE CENTRAL REGISTRIES OF OCCUPATIONAL AND MEDICAL EXPOSURE IN THE CZECH REPUBLIC

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### Abstract

This paper is intended to provide some insight into the recent situation in the Czech Republic concerning the registration and evaluation of occupational and medical radiation exposures. Since 1993 the creation of the Central (national) Registries of Occupational (CROE) and Medical Exposure (CRME) has been started. One of the main functions of these registries will be to provide statistics to guide policy making on a national basis. Authors pick up their presentation in previous national conference in Jachymov last year and continue with further detailed information on the structure of creating programs and discuss some actual arising problems.

### Introduction

The actions leading to the creation of CROE and CRME were opened in 1994 by Radiation Hygiene Centre of National Institute of Public Health under the financial participation of the Ministry of Industry and Trade and Ministry of Health. In accordance with the changes in the structure of radiation protection in our republic this year, registries are now created in new National Institute of Radiation Protection which is supervised by State Office of Nuclear Safety.

### Croe - Recent Situation

The basis of the registration and evaluation of OE is the individual monitoring of classified workers. The employer's duty, covered by the Regulation No.5 / 1979 in our country, is to secure individual monitoring of radiation workers and record results of monitoring.

At present there are five dosimetry services, about twenty thousand monitored workers and one thousand registered employers in CZ. Distribution of workers according to four basic occupational groups is given in Tab.1. As it has been already mentioned the creation of central registration system has been started since 1994 and following steps have been already done: the choice of the company for software covering of the system, the start of the co-operation with all our dosimetric services, methodology unification of OE dosimetric evaluation in our country (1).

**Tab.1:** Numbers of monitored workers (in thousands) for four basic occupational groups in the Czech Republic

| Occupational group             | Year  |       |       |       |       |
|--------------------------------|-------|-------|-------|-------|-------|
|                                | 1 975 | 1 980 | 1 985 | 1 990 | 1 993 |
| Uran.industry <sup>1)</sup>    | 3,5   | 10,3  | 10,3  | 7,9   | 3,3   |
| General industry <sup>2)</sup> | 11,2  | 5,4   | 6,8   | 7,8   | 5,7   |
| Medicine <sup>2)</sup>         | 3,7   | 5,6   | 7,1   | 8,2   | 11,1  |
| NPP <sup>3)</sup>              |       |       |       | 1,5   | 2,6   |

<sup>1)</sup> Dosimetry Service of Uranium Industry

<sup>2)</sup> National service of Personal Dosimetry, Ltd.

<sup>3)</sup> Dosimetry Service of NPP Dukovany

The database system is using ORACLE and operating in HP computers. The CROE databases are - personal, utilities (employers), dosimetric services, values of the personal dosimetric quantities, accidents, cumulative five years values.

Databases contain detailed identification of employers including their activity categories (Tab.2) and the dose records for all monitored workers with details of their age, sex, occupational category, type of handling radiation source. The data will be reorganized annually and individual dose assessment will be maintained for the current year and previous five years. Earlier data will be archived. The system uses special identification number for workers (birth number) and employers (random number) and all data are treated as confidential. Recently the registration cards serving for a contact between CROE and dosimetric services and employers are created. The cards will provide for CROE entrance data of all radiation workers in CZ and any changes in their registration.

The contact with the International System on Occupational Exposure was also opened up in 1994 and the created national system of ORE registration is built in the harmony with the recommendations and demands of this international system. This is a reason for instance for such detailed structure of occupational categories in NPP.

**Tab.2:** Employer's activity categories in CROE

|                                 |  |
|---------------------------------|--|
| 1.0. Health service             | 4.2. Chemical                            |
| 1.1. Hospitals                  | 4.3. Mining                              |
| 1.2. Other medical facilities   | 4.4. Building                            |
| 1.3. Special medical facilities | 5.0. Uranium industry                    |
| 2.0. Education, research        | 6.0. Defence                             |
| 3.0. Energetics                 | 7.0. Agriculture, food                   |
| 3.1. NPP Dukovany               | 8.0. Transport                           |
| 3.2. NPP Temelín                | 9.0. Specialized facility                |
| 3.3. Others                     | 9.1. Customs, Inspectorates, Supervision |
| 4.0. General industry           | 9.2. Services, Repair work, Tests        |
| 4.1. Engineering                |  |

The start of routine work of CROE is planned for next year. CROE will have a number of functions - to provide the new employer with summarized information of an individual's dose history, to guarantee a right annual dose calculation for workers with two or more employers, to provide statistics to national regulatory authorities.

#### **Crme - Recent Situation**

The survey and evaluation of medical radiation exposure (MRE) exists as a part of all reports of UNSCEAR from 1958. The aim of these world-wide studies is an estimation of world-wide dose, analyse of frequencies and dose distributions and determination of time trends in this area. This information enables us to evaluate the regional differences in the use of sources of ionizing radiation (SIR) in medicine and to determine topics of interest on this field. Concerning the problems with the collection of data there are several ways used in different countries - organization of national surveys, co-operation with hospitals, universities, health insurance companies (2, 3). Concerning the problem with the evaluation of MRE - one of possibilities is to take account of age and sex differences in risk coefficient and use a new

quantity for evaluation of detriment from MRE (4). Generally it is possible to say that taking account of age and sex of patients, a detriment is significantly decreasing. This estimation depends of course also on demographic structure of individual countries.

We are taking account of all these problems and we would like to avoid them creating the national registration system in our country. In CZ there are recently 350 radiodiagnostic, 52 nuclear medicine and 40 radiotherapy workplaces. There was performed about 9.5 mil radiodiagnostic, 250 ths nuclear medicine and 22 ths radiotherapy procedures (numbers from Institute of Health Information Systems of Ministry of Health of CZ, in 1993). This is big amount of data and it is impossible to sort all of them according to all demanded parameters.

There are three main sources of data :

- the regular annual statistic survey of Ministry of Health which is managed by Institute of Health Information Systems (IHIS), but there is no possibility to sort patients according to their age and sex , IHIS collects only the numbers of procedures and there is also problem with the clear definition of individual examinations,

- the organization of a national survey and use information systems of individual health utilities,

- there is problem with co-operation and organization of this survey,

- the co-operation with health insurance companies,

- there are twenty companies in CZ now, but one of them General Health Insurance Comp. (GHIC) is the biggest one which cover about 80% of our population.

**Tab.3:** Nuclear medicine, 1994, females and males

| females  | > 84 | 75-84 | 65-74 | 55-64 | 45-54 | 35-44 | 25-34 | 15-24 | 5-14  | 0-4 | total |
|----------|------|-------|-------|-------|-------|-------|-------|-------|-------|-----|-------|
| bone     | 85   | 600   | 1417  | 1071  | 960   | 213   | 69    | 138   | 154   | 42  | 4749  |
| renal    | 16   | 156   | 418   | 363   | 353   | 235   | 132   | 467   | 710   | 216 | 3066  |
| thyroid  | 28   | 93    | 254   | 207   | 313   | 169   | 133   | 67    | 4     | 1   | 1269  |
| liver+gb | 8    | 63    | 161   | 106   | 192   | 115   | 49    | 127   | 56    | 21  | 898   |
| brain    | 14   | 64    | 113   | 72    | 105   | 54    | 30    | 55    | 21    | 2   | 530   |
| lung     | 167  | 503   | 598   | 265   | 288   | 90    | 42    | 44    | 18    | 9   | 2024  |
| heart    | 3    | 14    | 64    | 73    | 163   | 100   | 33    | 66    | 35    | 5   | 556   |
| tomo sci | 4    | 20    | 120   | 120   | 181   | 60    | 13    | 10    | 0     | 3   | 531   |
| other    | 1    | 18    | 43    | 33    | 40    | 36    | 14    | 21    | 13    | 0   | 219   |
| total    | 326  | 1 531 | 3 188 | 2 310 | 2 595 | 1 072 | 515   | 995   | 1 011 | 299 |       |

| males    | > 84 | 75-84 | 65-74 | 55-64 | 45-54 | 35-44 | 25-34 | 15-24 | 5-14 | 0-4 | total |
|----------|------|-------|-------|-------|-------|-------|-------|-------|------|-----|-------|
| bone     | 93   | 526   | 1124  | 609   | 372   | 145   | 65    | 192   | 182  | 93  | 3401  |
| renal    | 33   | 143   | 359   | 228   | 283   | 155   | 160   | 616   | 525  | 305 | 2807  |
| thyroid  | 4    | 83    | 92    | 44    | 45    | 36    | 17    | 10    | 4    | 0   | 260   |
| liver+gb | 11   | 29    | 126   | 121   | 134   | 78    | 58    | 150   | 91   | 13  | 811   |
| brain    | 1    | 41    | 107   | 64    | 67    | 30    | 31    | 58    | 29   | 1   | 429   |
| lung     | 76   | 274   | 509   | 295   | 254   | 147   | 76    | 26    | 20   | 7   | 1684  |
| heart    | 0    | 15    | 82    | 112   | 170   | 72    | 27    | 98    | 81   | 0   | 657   |
| tomo sci | 4    | 24    | 148   | 172   | 223   | 88    | 27    | 4     | 0    | 0   | 690   |
| other    | 1    | 4     | 30    | 29    | 20    | 11    | 17    | 7     | 8    | 0   | 127   |
| total    | 223  | 1 064 | 2 577 | 1 674 | 1 568 | 762   | 478   | 1 161 | 940  | 419 |       |

Database GHIC obtains birth number of each patient from that it is possible to determine age and sex of patient and GHIC has unified list of all examination - so this is exceptional opportunity to obtain statistically significant data for MRE evaluation in our country. We will obtain only part of birth number of patient, the system will not operate with confidential personal data, it will be anonymous, but before this modification of data the system will identify each patient as individual - it means we will be able to say not only how many procedures were performed but also how many patients were examined. In principle we would like to collect data by both way in co-operation with GHIC and with selected representatives health utilities. The data from IHIS which are collected on the national level we can use for better approximation of collected data to a national level.

GHIC provided us with first data which are related to the region with 1,2 mil inhabitants (app. 10 mil inhabitants live in CZ now) in 1994. Partial data concerning a nuclear medicine are shown in Tab.3. The analyse and creation of methodology for regular collection of data is our main goal for near future.

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## THE EVALUATION OF OCCUPATIONAL OVEREXPOSURES IN THE CZECH REPUBLIC

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National Radiation Protection Institute (NIRP), following the word-wide trends, has started in 1993 the realisation of the Central Registry of Occupational Radiation Exposures (CROE). Since 1996 the CROE will be able not only to registrate individual doses for radiation workers in the Czech Republic, but also to estimate collective doses in different occupational categories, to determine and analyse time trends in these groups, to compare exposures for different practices with sources of ionizing radiation (SIR).

The accidental data-base for collection and dissemination information on unusual exposure events in national system of radiation protection will be established as the important part of CROE. The attention of CROE accidental data-base will be concentrated mainly on unintended events, including operating errors, equipment failures, or another mishap, consequences or potential consequences that cannot be ignored from the radiation protection point of view.

The content and form of the information entering to CROE accidental data-base is in principal specified in two parts:

- a cover sheet containing basic information about given overexposure (source of first information, time frame, type of radiation source, type of exposure, workers involved, etc.)
- a narrative account describing circumstances (exposure conditions, equipments, facilities, safety system, what happened), causes (inadequate design, construction, maintenance, written local rules, management, source security, infrastructure, information from the manufacturer, training, failure warning, safety systems, shielding, monitoring instruments, etc), consequences (medical factors, dosimetric evaluation, etc.) of the overexposure.

Record keeping of overexposures is based on two systems:

- if there is suspicion on overexposure at a workplace, licensee is obligated to estimate of the severity of given event; an investigation shall be conducted with the aim of identification of relevant circumstances and to assay and record the relevant doses and their distribution in the body. In the case when the annual limit of the effective dose equivalent ( $H_{E,L}$ ) is exceeded (for internal contamination 1/10 of the  $H_{E,L}$ ), the event must be reported to State Office for Nuclear Safety (its Regional Centers RC SONS). For serious incident (with possible medical consequences) NRPI is involved in evaluation of incident consequences;

- the second system is based on cooperation of CROE with dosimetric services; these services report directly to CROE immediately after the evaluation of dosimeters for given control period any excess of the  $H_{E,L}$  (or determined investigation levels). Thus identified events are then investigated by RC SONS directly at the workplace where the event occurred, and the analysis is made of whether a personal dose or false positive reading of the dosimeter is actually concerned.

183 persons were involved during the years 1954 - 1994 in 65 minor accidents and in indicated events detected in the Czech Republic (and former CSSR). 101 of them were exposed to external and 82 to internal radiation. Only 23 involved workers have had some health consequences (dermatitis with skin defects, cataracts). In some events surgical interventions - amputation of fingers, removal of a local deposit were used.

In the period 1975 - 1994, 106 dosemeter readings exceeding the value 50 mSv were reported by the second system ( $H_{EL}$ ). Following reinvestigation appeared that in most cases the primary evaluation was falsely positive; it was concluded that the values of  $H_{EL}$  were exceeded only in 45 of indicated findings.

Several, typical events are described in the presentation mainly from the standpoint of practical utilization for safer use of SIR. Also the analysis of health detriment ("cost") following from indicated overexposures may be important, mainly for the process of justification and optimization of the use of SIR in practice, and could lead to health detriment reduction.



## AIRCREW EXPOSURE TO COSMIC RADIATION. NEW ANALYSIS, RECOMMENDATIONS EURADOS<sup>1)</sup>

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ICRP 60 publication recommends to include aircrew members among occupationally exposed persons [1]. Many theoretical and experimental studies have been realized since 1991, several international bodies have taken part in discussion of topic. The progress achieved can be characterized in the following way, the most of results are based on studies and analysis realised in the laboratory of authors.

### 1. On-Earths\* Reference Fields

Cosmic radiation on the board of an aircraft consist of two components: directly ionizing radiation (electron, proton - low LET) and neutrons (high LET). Neither composition nor the energy spectrum of usual on-Earth calibration sources (<sup>60</sup>Co, <sup>252</sup>Cf) do not correspond to the field on a board. That is why high energy reference fields behind shielding high energy accelerators at CERN (and Dubna) have been created and intensely studied. Their typical characteristics following from the results of our measurements are presented in Table 1.

Many different measuring instruments and detectors have been tested in all these reference fields, it was proved that hard fields (top concrete at CERN) neutron (high LET) component is very convenient for on-board calibration. A typical comparison is shown in Table 2. One can see there that the remmeter NM2 as well as bubble damage neutron detectors (BDNDs) underestimate (about twice) H\*(10) on the board when calibrated with <sup>252</sup>Cf neutrons. This correction factor has been taken into account as well as the multiplication by 1.25 in the case of low LET measuring instruments [2].

**Tab. 1** Dosimetric characteristics of on-Earth high energy reference fields.

| Characteristics                  | Value per hour                 |            |                                      |            |
|----------------------------------|--------------------------------|------------|--------------------------------------|------------|
|                                  | CERN<br>at $7 \times 10^5$ PIC |            | Dubna<br>at $I_p \sim 1 \mu\text{A}$ |            |
|                                  | top iron                       | top concr. | soft field                           | hard field |
| $E_N$ ; MeV                      | 1,9                            | 49,8       | 0,253                                | 12,4       |
| $H^*(10)$ ; Sv.cm <sup>2</sup>   | 1,60-10                        | 2,80-10    | 2,90-11                              | 1,05-10    |
| $\Phi_{th,n}$ ; cm <sup>-2</sup> | 4,72 06                        | 8,25 05    | 1,1 08                               | 4,2 06     |
| Low LET $H^*(10)$<br>μSv         | 60                             | 68         | 790                                  | 130        |
| High LET $H^*(10)$<br>μSv        | 1350                           | 375        | 4600                                 | 1200       |

<sup>1)</sup> Work partially supported through the EC Project FI3P-CT92-0026

**Tab. 2** Comparison of direct readings of high LET instruments.

| Radiation field  |         | Directly read value of $H^*(10)^{1)}$ ,<br>$\mu\text{Sv}$ , by |         |                       |
|------------------|---------|--|---------|-----------------------|
|                  |         | NAUSICAA-TEPC  | M2      | BDNDs'                |
| Prague- Montreal | 9,5 km  | 1,24±0,32  | 1,0±0,1 | 1,3±0,2 <sup>2)</sup> |
| Prague           | 10,1 km | 2,38±0,25  | 1,3±0,1 | 1,4±0,2 <sup>3)</sup> |
| A 310-300        | 10,7 km | 3,98±0,42  | 1,6±0,2 | 1,3±0,2 <sup>2)</sup> |
| February 1995    | 11,3 km | 3,35±0,42  | 1,8±0,2 | 1,4±0,2 <sup>3)</sup> |
| CERN             | T6      | 524±25   | 267±7   | 270±0,20              |
| top concrete     | S2      | 644±60   | 303±7   | 310±20                |

<sup>1)</sup> per hour on board, per  $10^6$  PIC counts at CERN

<sup>2)</sup> Average value for 9.5 and 10.7 km

<sup>3)</sup> Average value for 10.1 and 11.3 km

## 2. Exposure levels on the board

### 2.1. In-flight measurements

In-flight measurements on the board of commercial aircraft have been since 1991 realized during about 20 flights, seven of them were typical long-haul ones. Flight routes extended from the 1.3 °N up to about 65 °N, flying altitudes varied from 8.2 km to 12.5 km. Basic conclusions from these studies can be formulated in the following way:

a) The readings do not depend on the position in the aircraft ( $1\sigma_{\text{rel}} \pm 10\%$ ); a good agreement of results obtained with different instruments and detectors has been stated.

b) They decrease when going to the equator, mostly between 50 °N (4 GV of geomagn. rigidity) and 20 °N (11 GV), being in the equator region about two to three times lower for directly ionizing component, more than four times lower for neutron component.

c) They increase with flight altitude, the average increase for each 600 m (2000 feet) is for northern ( 50 °N) region and both components about 15%.

d) Following the actual conditions of flight (altitude, wind, etc.) the integral value of dose equivalent can differ for the same route up to the factor of 1.5.

e) The level of exposure can be estimated at 50 °N, at flight altitudes about 11 km and the period since the end of 1992 to about 6 to 8  $\mu\text{Sv}$  per hour.

### 2.2. Solar cycle influence on the exposure level

The exposure level due to galactic cosmic radiation (predominant contribution for the most of time) is inversely proportional to the solar activity. The most of our measurements has been performed since the beginning of 1993, when cosmic radiation level on the base of data from Lomnický štít and/or Calgary stations vary only a little ( $95 \pm 3\%$ ) of reference value. Only data with substantially lower countings had been obtained at the end of April 1991. The relative ratio of monitors' countings is  $1.14 \pm 0.02$ , the relative readings are equal to  $1.16 \pm 0.02$ . These ratios are, however, comparable with uncertainties of measurements. It should be also mentioned that very energetic solar flare had place at 29th September 1989. Daily average values increased by 20% at Lomnický štít (4 GV), by 80% at Calgary (1 GV).

## 3. Radiation protection aspects

a) The usual limits of annual aircrew flight hours correspond at 11.3 km to about 4 mSv per year, with new ICRP 60 conversion factors to about 5 mSv per year. It is well bellow the limit, higher than for other groups of occupationally exposed persons (bellow 1 mSv in medicine, resp. 1.6 mSv in industry [3]).

b) Monthly flight hours limit does not exclude that the exposure of a pregnant women can exceed 1 mSv during this period.

c) The aircrew exposure should therefore be checked, controlled and administered as conscientiously as for any other group of occupationally exposed persons.

#### **4. European activities**

In the collaboration between DGXI and DGXII of CEC two main activities have been started since 1992:

a) CEC Project has been formulated, the studies in the frame of it have continued up to the end of September 1995. Many results like these presented in this work have been acquired.

b) A Working group 11 of EURADOS "Exposure of Air Crew to Cosmic Radiation" has been formed to prepare basic analysis and recommendations concerning the topics. The basic recommendations are the following [4]:

- air crew flying routinely at altitudes over 8 km are deemed to be category B workers, it is therefore important to estimate, record, control and, where necessary, to limit the doses;

- the preferred procedure in order to estimate doses to air crew or frequent flyers is to determine route doses and fold these data with data on staff rostering;

- where doses may exceed the limit for category B workers (6 mSv per year), on-board monitoring of dose may be appropriate.

- an equivalent-dose limit of 1 mSv for the embryo and foetus should be specify for air crew.

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## TO THE USE OF BUBBLE DETECTORS IN PERSONAL NEUTRON DOSIMETRY<sup>1)</sup>

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ICRP 60 recommendations require to improve the lowest limit of detection (LLD) of an individual neutron dosimeter by a factor estimated up to about five [1]. Bubble neutron detectors could fulfill such conditions, the LLD of systems already available is claimed to be well below 10  $\mu\text{Sv}$ . We have tried to test them at different neutron sources. The results obtained are presented.

### 1. Bubbles Detectors Tested

Two types of bubble detectors are commercially available: bubble damage neutron detectors (BDNDs\*) from Bubble Technology Industries, Chalk River; and superheated drop detectors (SDDs\*) from APFEL Industries, New Haven.

BDNDs\* tested had the sensitivity about 1 bubble per 1 Sv of  $H^*(10)$  of AmBe neutrons, they were evaluated by eye counting (20 to 30 bubbles per detector). Two types of reusable BDNDs\* were tested: BD-100R without and PND with temperature compensation, both with neutron energy threshold about 100 keV.

SDDs\* tested had the sensitivity about 3 bubbles per 1  $\mu\text{Sv}$  of  $H^*(10)$  from AmBe neutrons, they were evaluated using APFEL Survey Meter Model 202. SDDs\* with three different energy thresholds have been used: 0.1, 1 and 6 MeV.

### 2. Results

#### 2.1. Counting statistics; LLD

Multiple irradiation of several pieces of BDNDs\* (type PND) was realized with AmBe neutron source. The standard deviation of a single reading was found to be regularly close to  $\sqrt{N}/N$  ( $N$  = number of bubbles). The bubbles appearance with the exposure follows therefore the normal distribution. The precision  $\pm 30\%$  is hence achieved when total number of bubbles is above 10. At the sensitivity 1  $\mu$  bubbles per 1  $\mu\text{Sv}$ , 10  $\mu\text{Sv}$  are measurable with mentioned precision. With increasing sensitivity, the LLD decreases.

#### 2.2. Energetical dependence of BDNDs\*

Energetical dependences of bubble detectors have been studied in neutron beams and fields characterized in Table 1. The relative responses of BDNDs\* established in the first run of calibrations (1993-1994) are presented in Table 2.

At the end of 1994 we were able to purchase a new lots of BDNDs\* of both mentioned types and to repeat the most of calibration. The results of this second run are presented in Table 3.

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<sup>1)</sup>Work partially supported through EC Project FI3P-CT93-0072

**Tab. 1** Neutron field for energetical dependency studies of bubble detectors.

| Neutron source                        | $E_N$ [MeV] | $H^*(10)^{1)}$ [Sv.cm <sup>2</sup> ] | Neutron source              | $E_N$ [MeV] | $H^*(10)$ [Sv.cm <sup>2</sup> ] |
|---------------------------------------|-------------|--------------------------------------|-----------------------------|-------------|---------------------------------|
| SIGMA                                 | 0,1         | 2,29 -11                             | AmBe                        | 4,4         | 3,80 -10                        |
| CANEL+(H <sub>2</sub> O)              | 0,096       | 3,47 -11                             | PuBe                        | 4,2         | 3,80 -10                        |
| CANEL+                                | 0,19        | 7,26 -11                             | JINR-soft <sup>2)</sup>     | 0,25        | 2,90 -11                        |
| <sup>252</sup> Cf/D <sub>2</sub> O/Cd | 0,54        | 9,10 -11                             | CERN-iron <sup>3)</sup>     | 1,9         | 1,60 -10                        |
| AmF                                   | 1,5         | 3,40 -10                             | JINR-hard <sup>2)</sup>     | 12,5        | 1,05 -10                        |
| <sup>252</sup> Cf                     | 2,1         | 3,40 -10                             | CERN-concrete <sup>3)</sup> | 49,8        | 2,80 -10                        |

<sup>1)</sup> ICRP 26 conversion factors

**Tab. 2** Relative responses of BDNDs\* (BD 100R, 0.8 or 2.2 bubbles per 1  $\mu$ Sv of  $H^*(10)$  to neutrons of different sources - 1st run.

| Neutron source                        | Relative response | Neutron source | Relative response |
|---------------------------------------|-------------------|----------------|-------------------|
| SIGMA                                 | 0,48±0,06         | AmBe           | 0,99±0,03         |
| CANEL+(H <sub>2</sub> O)              | 0,89±0,11         | PuBe           | 0,97±0,06         |
| CANEL+                                | 1,06±0,07         | JINR-soft      | 0,95±0,09         |
| <sup>252</sup> Cf/D <sub>2</sub> O/Cd | 1,01±0,06         | CERN-iron      | 0,98±0,05         |
| AmF                                   | 1,06±0,07         | JINR-hard      | 0,69±0,08         |
| <sup>252</sup> Cf                     | 1,11±0,08         | CERN-concrete  | 0,60±0,04         |

**Tab. 3** Relative responses of BDNDs\* (BD 100R and PND, 1 bubble per 1  $\mu$ Sv of  $H^*(10)$  to neutrons of different sources - 2nd run.

| Neutron source                        | Relative response |           |
|---------------------------------------|-------------------|-----------|
|                                       | BD-100R           | PND       |
| SIGMA                                 | 0,52±0,04         | 0,54±0,04 |
| CANEL+(H <sub>2</sub> O)              | 1,13±0,09         | 1,06±0,08 |
| CANEL+                                | 1,22±0,29         | 1,06±0,08 |
| <sup>252</sup> Cf/D <sub>2</sub> O/Cd |                   |           |
| AmF                                   | 1,10±0,08         | 1,16±0,08 |
| <sup>252</sup> Cf                     | 1,34±0,14         | 1,38±0,15 |
| AmBe                                  | 1,00±0,06         | 0,94±0,07 |
| PuBe                                  | 1,05±0,07         | 1,03±0,07 |
| JINR-soft                             |                   | 1,13±0,13 |
| CERN-iron                             | 1,28±0,13         | 1,17±0,10 |
| JINR-hard                             |                   | 0,70±0,07 |
| CERN-concrete                         | 0,63±0,04         | 0,62±0,04 |

General conclusions from both Tables can be formulated in the following way:

a) With the exception of thermal neutron source SIGMA (50% of  $H^*(10)$  from thermal neutrons) and high energy reference fields there is a reasonable agreement of data measured with BDNDs\* and expected values.

b) Nevertheless, the new lots seem to have a little different energetic dependence. The relative responses for "soft" fields are for them systematically higher than for previous samples.

Apparently, the response to energies between 0.01 and 1 MeV is for these new lots relatively higher.

c) The underestimation of high energy neutrons is typical for any LET-threshold type detectors [4]. It should be kept in mind when BDNDs\* are used as dosimeters in high energy neutron environment (aircraft board!).

### 2.3. Energetical dependence of SDDs\*

The results of first run studies of the energetical dependence of SDDs\* are presented in Table 4. One can see there that:

**Tab. 4** Relative responses of SDDs\* to neutrons of different sources

| Neutron source    | Relative response of SDD with the threshold |             |             |
|-------------------|---|-------------|-------------|
|                   | 0.1 MeV                                     | 1 MeV       | 6 MeV       |
| AmBe              | 1,00 <sup>1)</sup>                          | 0,48±0,06   | 0,16±0,02   |
| AmF               | 1,03±0,15                                   | 0,028±0,004 | 0,028±0,005 |
| <sup>252</sup> Cf | 1,20±0,15                                   | 0,20±0,03   | 0,09±0,01   |
| PuBe              | 0,94±0,08                                   | 0,40±0,05   | 0,16±0,02   |
| CERN-top iron     | 0,70±0,10                                   | 0,09±0,02   | 0,17±0,03   |
| CERN-top concrete | 0,50±0,05                                   |             |             |

<sup>1)</sup> Reference value

a) The energetical dependence of SDD 100 is comparable with the dependencies of BD-100R and PND, the underestimation of high energy neutrons included.

b) The use of SDD with different energy thresholds can provide interesting spectrometric information; the sources like <sup>252</sup>Cf can be clearly distinguish from AmBe when comparing the responses of SDD 100 and SDD 6000 (1000).

It should be also mentioned that the spectrometry can be also realized with sets of BDNDs\*. We have tested such a set (6 detectors with thresholds from 0.01 to 10 MeV) in hard and soft field at JINR Dubna with quite satisfactory results [5].

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**PERFORMANCE OF NECESSARY INVESTIGATION ON  
WORKPLACES WITH IONIZING RADIATION IN NEW  
ECONOMICAL CONDITIONS**

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**Abstract**

In the presentation the attention is given to the obligation of license possessor for the operations with ionizing sources to perform not only the measures from point of view of radiation protection against ionizing radiation (it was and it is obligatory for all possessor of license) but also to ensure Quality Assurance. This measurement may perform only the institutions and persons to whom the approval were given from the state supervisor bodies. The proposal of the system of licensing of qualified persons to this activities is presented.



## **THE PRESENT PROBLEMS OF HYGIENIC SUPERVISING OF WORKPLACES WITH IONIZING RADIATION SOURCES**

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Important for population health protection is both risks monitoring of artificial and natural sources of ionizing radiation and hygienic supervising of the workplaces, where this kind of radiation is used. In Slovakia is supervising exerted by Departments of Health Protection for Radiation, which are established on four State Health Institutes. They are directed under the law 272/1994 through General hygienic of Ministry of Health.

These departments deal not only with supervising of these workplaces, but in this article is described only this problem and especially in the State Health Institute of Slovak Republic Capital Bratislava. Compared with other three departments of State Health Institutes the activities of this one are specific. As first, in Bratislava are supervised all kinds of workplaces with ionizing radiation sources, that in the rest of Slovakia occur, with the only exception from an operation of a nuclear reactor. Specific conditions consist in fact, that there are concentrated relatively great many various sources in relatively small area with high residential density. As second, in Bratislava are concentrated research and school facilities, central authorities and a lot of specific workplaces, where ionizing radiation sources are used. There is contents of work often changed at these workplaces and consequently these workplaces are often established, respectively disestablished.

From briefly characteristic of specific conditions in Bratislava follows, that not all problems related to supervision workplaces with ionizing radiation sources can be solved on routine basis. Most problems consist in the present economic transformation of State Corporations. Abolishing of previous State Corporations and arising of new organizations means new field of their activities. It often happens, that previously used ionizing radiation sources, X-ray tubes, or radioactive sources, are not longer to use and it is necessary to remove corresponding workplaces. Substantially it should not be a problem, because each of these workplaces has an estimated supervisor who should be able to manage new situation right. But the reality is often another one. Problems related to liquidation of workplace with ionizing radiation sources are only of margin interest for management of liquidated corporation. In addition, the supervisor and his chief are not already employed in the new organization. This state is found by routine hygienic inspection. In such case a matter of importance is to find what has happened with ionizing radiation sources. If State Corporation possessed only X-ray tubes, it does not come to great problems. Mostly X-ray tubes are provably disassembled, or even destroyed. In the case of radioactive sources a situation is more complicated. The radioactive source is mostly stored according to the regulations and it can be also found his certificate and documentation of the workplace. The cardinal problem is to find someone, who has known former workplace, because supervisor and chief department are not already employed in a new organization. In spite of this, the problem can be solved within short time under a condition of good cooperation between new organization management and the representative of State Health Institute. To illustrate the solving of one of many such problems follow an example.

The big State Corporation with series of subsidiaries in whole Slovakia was divided to many new smaller Joint-stock Corporations. A subsidiary in Bratislava possessed workplace with X-ray tube and sealed radioactive source of medium radioactivity. During a routine hygienic

inspection was found, that the original establishment was abolished, all personnel dismissed and another organization is going to move at this place. New organization personnel has not known, that the previous workplace was such one with ionizing radiation sources. Fortunately, the immediately made measurements proved no presence of higher radiation on this place. So it was necessary to obtain the succeeding organization adress. It was no problem, but it turned of, that it is outside of Bratislava. Then followed proceedings with succeeding organization management, where explanation was asked what has happened with X-ray tube and radioactive source. It was found, that the new management has no information about it and it was in position, it is not a matter of new organization. The situation was complicated by the fact, that new management had no connection to previous personnel and had not sufficient information about abolished establishment. Therefore firstly it was necessary to provide basic information about risks connected with loss of radioactive source, further about regulations of this branch and consequences of breaking them. At last there were stated duties for succeeding organization with respect to this problem and it was given a term to solve it. After relatively short time our department was informed, that in archives were found documents of selling X-ray tube and radioactive source to another organization. In this point the problem was solved and it could be proceeded in routine procedure. Firstly, it was established workplace with ionizing radiation sources complying to regulations by new owner, incidentally again in Bratislava. At last the workplace and approve to treat with ionizing radiation sources on original place could be abolished.

Concernig this example let me allow following comment. For this specific case it was probably useful campaign against nuclear energetics, because as a result of it a general public obtained basic information about radioactive sources and about their possible hazard to human health. In this way arised respect, probably even fear of radioactive radiation, that helped to solve beforehand meant problem.

Described problem of supervising workplaces with ionizing radiation sources was not the only one, but there had to be solved more similar, more or less complicated cases. It is evident, that there were broken corresponding regulations by organizations and it should never happen. In fact, especially at present time, it is in other way and it is in charge State Health Institutes to supervise such workplaces and if necessary, to ensure solving of arised problems. The routine course, namely inspection, finding defaults, directing to put them right, to make record, apply sanctions, is possible, but it takes a lot of time. I mean the better way consists in quick, uformal action, with first rate aim to ensure qualified supervision and treating with radioactive source. The reason of this opinion is in experience and in cases described by literature. Unqualified treating with radioactive sources ends sooner or later with some accident which has more serious consequences than it is in the case of unexpected accident by qualified treating.

At present time there are a lot of problems, that Department of Health Protection for Radiation deals with. Many of them is necessary to solve by less usual way. I consider useful to emphasize this one described in this article. Its underestimation would lead to undesirable serious consequences, especially in Bratislava.



## CEMA AND OTHER NEWS IN QUANTITIES

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### **Summary**

Information is provided on the newly by ICRU introduced quantity CEMA defined as the energy per unit mass lost by the charged particles produced by uncharged particles or from a primary charged particle beam. Thus CEMA and reduced CEMA, not including the energy transported by delta rays, are intermediate quantities between the fluence and kerma and the absorbed dose. There are also some changes in names and definitions of some other radiation and dosimetry quantities recommended in recent years by ICRU.



## THE NEW SAFETY FUNDAMENTALS FOR RADIATION PROTECTION

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### **Summary**

Information is provided on the Safety Fundamentals - Radiation Protection and the Safety of Radiation Sources adopted on 15. June 1995 by the Board of Governors of IAEA as a document superior to the Basic Safety Standards and other derived IAEA documentations. To the three basic principles - justification, limitation and optimisation the fourth - safety of sources is added.



## THE CALIBRATION METHOD FOR PERSONAL DOSIMETRY SYSTEM IN PHOTON AND NEUTRON RADIATION FIELDS

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### Abstract

The main objectives of individual monitoring are to obtain informations on effective doses  $E$  in significantly irradiated organs and tissues. The ICRP in the Publication No.60 [1] recommends that the dose limitation for stochastic effects should be based on the quantity annual effective dose. As the quantity  $E$  is not directly measurable, operational quantities are personal dose equivalents  $H_p(d)$  in the depth  $d$  of 0,07 mm and 10 mm are conducted. This operational quantities together with the energy of radiation serve for assessment of  $E$ .

In operational praxis of individual monitoring of photon and electron radiation and thermoluminescent dosimeters are used. For personal neutron dosimetry solid state track detectors in contact with fissionable radiators U Al and Th are applied for personnel neutron dosimetry.

The reliability of interpretation of individual monitoring depends on the accurate calibration of the whole dosimetry system. From the practical point of view we distinguish between type testing procedure which involves the testing of the performance characteristics of the system as a whole and routine calibration under a given set of radiation and environmental conditions. The type testing of our dosimetry system is performed with standard photon radiation fields within the energy range 15 keV to 1.25 MeV and electron radiation fields within the range 0.2 MeV to 3 MeV. For type testing of neutron dosimeters  $^{252}\text{Cf}$  and  $^{241}\text{Am-Be}$  radionuclide neutron sources are used, as well as a 14 MeV neutron generator. The neutron sources moderated by various moderating and absorbing materials are also used.

The routine calibration of individual photon dosimeters is carried out using a  $^{137}\text{Cs}$  calibration source in the air kerma quality in the dose range 0,2 mGy to 6 Gy. For the calibration purposes a special device was constructed, containing 3 sources of  $^{137}\text{Cs}$  with various activities located in a PbW container. For ensuring the accurate and reproducible calibration of dosimeters, the irradiation in the open air is carried out on a plexiglas ring (10 mm thick and 30 mm width) at 30 cm distance between the source and dosimeter.

The moving of radiation source into the exposure position is done by a magnet in a brass leading tube, controlled by an electronic timer. A set of ionization chambers (Victoreen) with indications calibrated against primary standards are used for measurements. Calibration of film badge is also performed in standard X-ray field of 45 keV energy (70 kV + 0,5 mm Cu and total 1,0 Al) realised by Seifert rontgen aparaties in the State Institute for Publish Health in Prag. The measured quantity is also air kerma in the range of 0,1 mGy to 100 mGy. The doses in the calibration beam are measured with Victoreen and Ionex Dose Master chambers. In the cases the traceability of calibration beams were tested in different laboratories within the intercomparison organized by IAEA Vienna.

The type testing procedure is based on the use of the ICRU tissue equivalent 30x30x15 cm PMMA slab phantom. The required energy and angular response is determined from the calculation of equivalent dose distributions in the PMMA slab phantom. The results are used to relate the response required for  $H_p(10)$  and  $H_p(0,07)$  to that required for air kerma, by means of

set of conversion coefficients, published in BSS 1995. Dosemeters are irradiated on the phantom in an energy interval 10 keV to 1,25 MeV.

The routine calibration of neutron dosemeters is realized by  $^{252}\text{Cf}$  fission spectrum in the 50 cm distance. The dosemeters are located on the PMMA ring (1 cm thick and 3 cm width). The dose equivalents were estimated using the values of emission of neutrons from manganese bath. The type testing of neutron dosemeters was performed in collaboration with Neuherberg laboratory on neutron generator with neutron energies of 0,57; 1,9; 5,3 and 15,1 MeV.

For this purpose the fission neutrons moderated with 40 cm of Pb and 20 cm of Pb were also used. The dose equivalent measurement was performed on the basis of spectra measurements by Bonner Spectrometer.

The fading and angular dependence testing is also included in the tests of both dosemeter systems.



## DERIVED INTERVENTION LEVELS IN EARLY STAGE OF NUCLEAR ACCIDENT DEVELOPMENT

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Measures for protection of health and property of public in the case of nuclear accident are based on optimal application of so called intervention levels. Optimal application of intervention levels requires the structure of decision scheme elaborated in advance. The actual flow of decision depends on:

- prognoses of mathematical modelling of possible course of nuclear accident, and
- results of monitoring of radiation situation.

In the case of nuclear accident the data from these two sources are mutually complementar. The aim of this contribution is to analyze their mutual cooperation and to suggest such procedure of monitoring or radiation situation which could be used for suggestion of protective measures.

In this contribution we aim exclusively to specify the zones of protection planning in the accident place surrounding for these urgent measures:

- regulation of free movement of persons,
- sheltering,
- iodine prophylaxy,
- temporary evacuation,
- long term or permanent emigration.

At the specification of zones of planned protection it is also coming out that regulation of movement of persons, sheltering and iodine prophylaxy were ordered in advance based on the evaluation of the crashed establishment state.

In such situation the decision on protective measures in the time interval 6 to 12 hours after the beginning of accidental release is forwarding mainly to:

- withdrawing the accepted orders on measures,
- transition from sheltering to temporary evacuation

### **The criterion for temporary evacuation is:**

- probability of exceeding the effective dose 100 mSv for children up to 10 years of age and pregnant women and 500 mSv for other population within 48 hours after beginning of accidental release,
- probability of averting the effective dose 50 mSv up to 7 days, 100 mSv up to 15 days and 150 mSv up to 30 days for all population groups.

Modelling of radiation accident development should in its output give the size of planned protection zones to organize the temporary evacuation in order to fulfil the criteria quoted above.

According to the first results of model prognoses it is carrying out the monitoring of actual radiation situation in the time range 6 to 12 hours. During organization of monitoring it should be kept in mind the fact, that the real evaluation of accidental release consequences can be performed only after the release has stopped or after its prevailing part had escaped. Decision based on monitoring of radiation situation within duration of release can considerably underestimate the final situation.

What should be the contents of monitoring? It can be said in general, that the contents of monitoring should be these dosimetric data which may be interpreted by means of a proper model as intervention level. There are mainly:

- kerma dose rate in air expressed as  $\text{mGy}\cdot\text{h}^{-1}$ ,
- equivalent dose rate in air caused by contaminated surface, expressed as  $H_x$  or  $H'(10)$ ,
- relative radionuclide composition of contamination of free surfaces expressed in Bq per sample,
- relative radionuclide composition of contamination of air expressed in Bq per sample.

#### Interpretation of values of kerma dose rate in air

Analyses of possible consequences of various types of accidents of nuclear power plants of VVER 213 and 230 type show that kerma dose rate in air within the first hours after escaping the prevailing fraction of accidental release can be interpreted as follows:

**Tab. 1** Interpretation of kerma dose rate in air after accidental release (distance from source 5 km)

| Duration<br>hod | $H_e$<br>[mSv/Gy.h <sup>-1</sup> ] |
|-----------------|------------------------------------|
| 48              | 30 - 110                           |
| 168             | 45 - 125                           |
| 732             | 75 - 170                           |

Norm value of effective dose is expressed as a sum of assumed contributions of irradiation from the passing plume, from inhalation and from contaminated free surfaces. The irradiation of skin and inhalation of resuspended activity had not been taken into considered. The range of effective dose presented in Table 1 is caused mainly by the size of escape, of its radionuclide composition and by time course of the proper escape.

It can be said in general that lower values of norm effective dose can be expected at accidental release with lower portion of rare gases and vice versa. These values of normed effective dose were calculated for the case of irradiation up to 40 hours, in distance 5 km:

**Tab. 2** Norm values of effective dose at 48 hours [mSv/mGy.h<sup>-1</sup>]

| Portion of NG<br>in release | 5 km   | 15 km  | 30 km |
|-----------------------------|--------|--------|-------|
| 0,85                        | 35-40  | 40-45  | 45-50 |
| 0,85-0,98                   | 60-65  | 60-65  | 60-65 |
| 0,995                       | 70-110 | 65-100 | 60-90 |

Values of norm effective dose had been calculated for the distance 5, 15 and 30 km from the source, and for irradiation time 48, 168 and 732 hours respectively after beginning of escape.

#### Determination of the size of planned protection zones

For the determination of size of planned protection zones had been used the expected values of effective dose at 48, 168 and 732 hours after beginning of accidental escape in the distance of 5, 15 and 30 km from the source. Calculation is performed under assumption that

the dilution factor in air has the value  $1.0 \text{ s.m}^{-3}$ . The dimension of expected effective dose was therefore  $[\text{mSv/s.m}^{-3}]$ . Then the ratio

$$\text{DIL}(\text{s.m}^{-3}) = \frac{\text{IL}}{\text{NH}_e}$$

where: IL - intervention level [mSv]

$\text{NH}_e$  - expected value of effective dose  $[\text{mSv/s.m}^{-3}]$

can be used for the assessment of the length of planned protection zone for arbitrary distance and weather type.

Calculated values of expected effective dose  $\text{NH}_e$  are presented in Table 3.

**Tab. 3** Values of dil for expected dose 100 mSv

| Duration of irradiation [hours] | DIL $[\text{s.m}^{-3}]$ |           |
|---------------------------------|-------------------------|-----------|
|                                 | MIN.                    | MAX.      |
| 48                              | 3.48 E -4               | 4.28 E -8 |
| 168                             | 2.34 E -5               | 3.07 E -8 |
| 732                             | 1.23 E -5               | 1.88 E -8 |

Values  $\text{NH}_e$  calculated for various distances from the source had differed only negligibly. It was possible however to estimate the regression dependence of DIL calculated for IL = 500 mSv and of total release. The regression equation has the shape

$$\text{DIL} = 7.74 \text{ E}+10 \times \text{Bq}^{-0.93}$$

Calculations for other types of accidental release are in progress.



## **RADIOACTIVITY MONITORING NETWORK OF SLOVAK HYDROMETEOROLOGICAL INSTITUTE AND ITS ACTIVITY WITHIN THE FRAMEWORK OF NUCLEAR EMERGENCY INFORMATION SYSTEM**

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### **Abstract**

SHMI radioactivity monitoring network is a part of nuclear radiation early warning system. This paper describes the aim and the structure of the monitoring system.

### **Introduction**

Nuclear power plant operation is joined with potential risk of people impact by accidental radioactivity releases. Powerful information systems are the sources of information of environmental radioactivity. They are the database for improving of effective countermeasures. Radioactivity monitoring network of SHMI performs the tasks of early warning system.

### **Aims of system**

The system from the beginning was made by six measurement places. They were equipped by dose rate equipment NB 3201. The effort of area monitoring network building led to the extending of monitoring places into 26 ones. They have been equipped by proportional detectors of FAG FHZ 621B.

In 1992 according to the cooperation between Czechoslovak Federative Republic and Federative Republik of German the integrated monitoring information system for radioactivity IMIS was performed. In the first stage of system building the user software was realised by firma Dornier. The workstation DEC 5000-240 with software was installed at SHMI. Also the staff was trained and the system was connected to the public data network Eurotel. In the second stage the measurement places of radioactivity monitoring network were connected to the system. In the framework of the information system Slovak Centre of Radioactivity Monitoring Network is superior to the SHMI informaticon system. It collects data from many organisations and also from SHMI monitoring network. According that fact the more efficient computer was installed in Slovak Centre of Radioactivity Monitoring Network. The computer DEC 5000-133 with software for radioactivity monitoring data collection was installed to SHMI. Fig.1 shows the transmissions of IMIS informations.

According to Agreement of Slovak Ministry of Environment and Austrian Federal Ministry of Health and Consumer Protection signed in may of 1994 data exchange from early warning systems have been made. For that purpose National Early Warning Centre of Vienna and National Communication Centre of SHMI of Bratislava Airport were connected by leased line. The data of Slovak measuring system are available as 10-minutes values and daily average values in the telecommunication centre at Bratislava Airport, from wich the data are transferred by local network (TCP/IP) to any partner-computer. The data of Austrian measuring system are available as 5-minutes average values, average values per hour and daily average values on the process control computer in the National Early Warning Centre in Vienna. Fig.2 shows the connection of parts of the system.

Measurement places are the information source of radioactivity monitoring network. Software preprocesses radioactivity data within meteorological data and transferes them to

telecommunication centre. Software for cooperation with dose rate detector works in two regimes:

1. automatic operation
2. manual operation

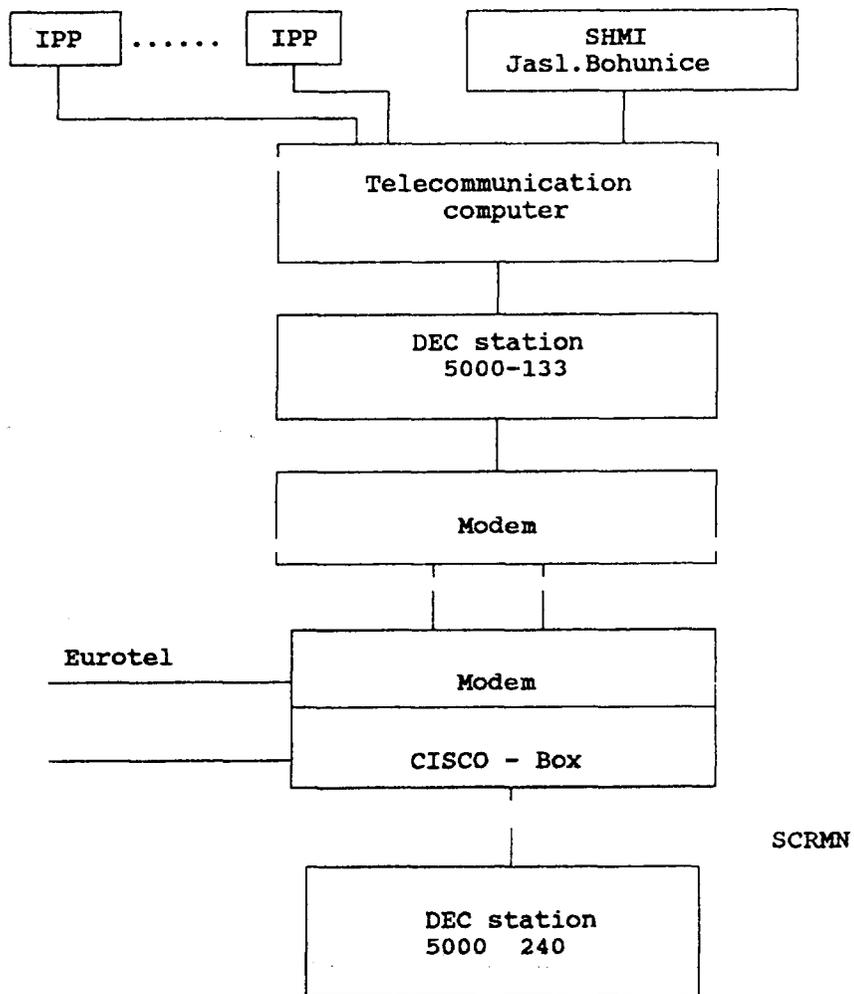
Stages of automatic operation:

1. normal operation (gama dose rate is lower then 500 nSv/hour)
2. increased radioactivity (gama dose rate is higher then 500 nSv/hour)
3. failure

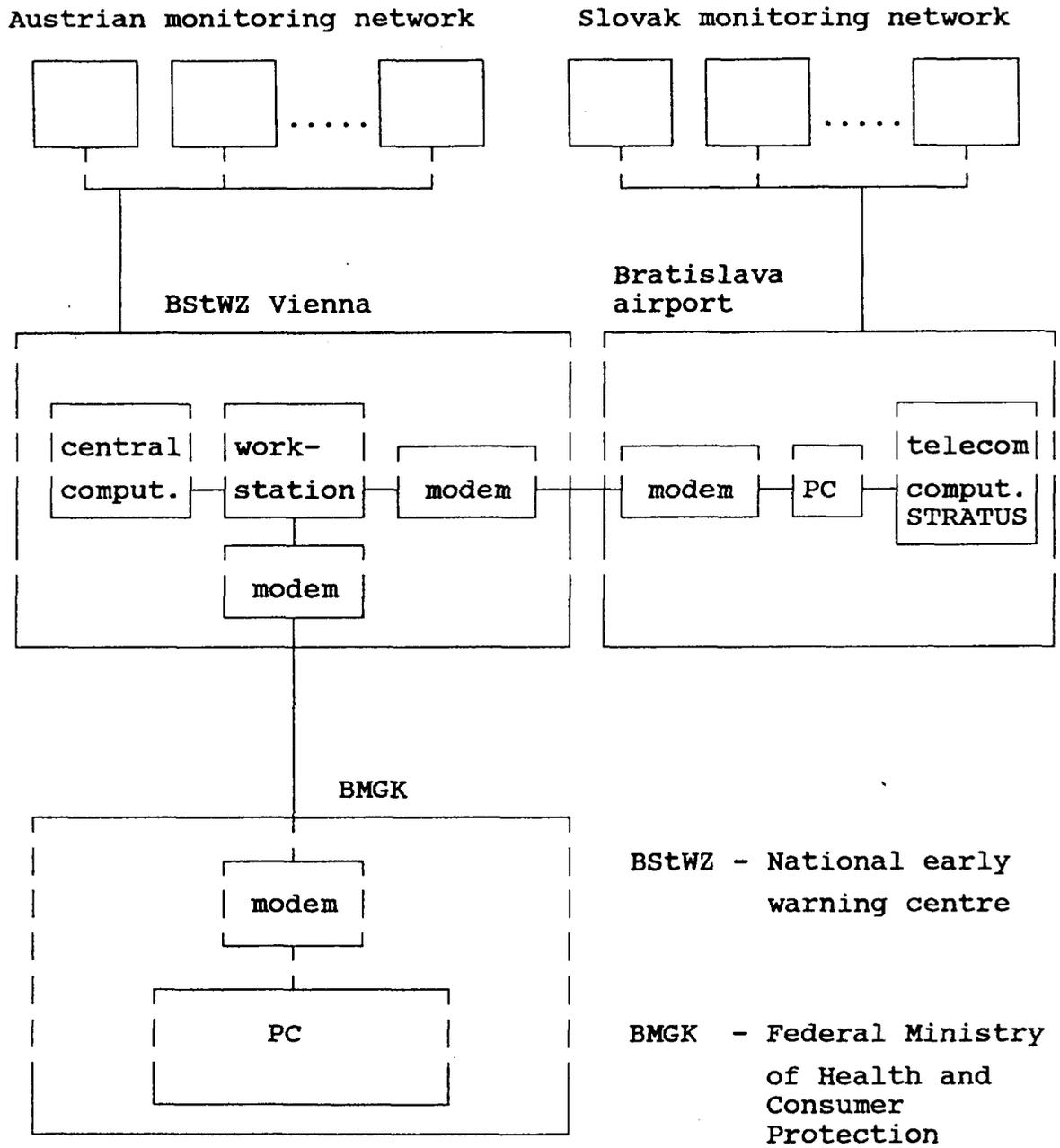
For information system IMIS in "normal operation" daily average values are transferred. In "increased radioactivity" two-hours average values are transferred.

**Conclusion**

This paper presents a short description of radioactivity monitoring network of SHMI and its connection with Austrian and German systems. It provides national means for the monitoring of the radiological effects of nuclear accident and for informing goverment departments and the public.



**Fig.1** Scheme of IMIS information system



**Fig.2** Connection of Austrian and Slovak early warning systems

## MOBILE UNITS FOR MONITORING OF RADIATION SITUATION

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### Abstract

The Chernobyl accident has clearly demonstrated the importance of establishing environmental monitoring systems for obtaining rapid information in accidental situation on the radiological conditions at the affected area and providing suitable data to the competent authorities for decision making. Mobile units suitable for monitoring of radiological impact at any selected location in the field play an important role in emergency preparedness for various types of nuclear accidents:

1. A major accident in a domestic nuclear power plant.
2. Accidents in nuclear power plants abroad especially in neighbouring countries. To this category should be added the risk from military nuclear power accidents.
3. Re-entry of nuclear powered satellites. The probability is small but it can not be excluded. This type of accident would result in highly radioactive fragments and particles that would have to be located.
4. Accidents when transporting radiation sources.
5. Illegal handling of radioactive sources and nuclear material.

Mobile units can be used for the measurement of dose and dose rate in wide range, ground surface contamination, nuclide specific activity concentration in air (with special attention to iodine in aerosol) as well as in other samples like soil, water, vegetation, food-stuff etc.

For the Radiation Monitoring Network of the Czech Republic a prototype of the small system for dose rate measurements and nuclide activity estimates suitable for use in mobile units was designed and tested in National Radiation Protection Institute.

The system consists of HPGe detector, multichannel analyzer, high pressure ionization chamber or proportional counter and portable computer working in multitask mode for storing and evaluating of the spectra as well as for dose rate data handling, recording, storing and searching and presentation. The information about actual position is provided by the global positioning system (GPS) Garmin on-line connected to the computer. This information is used later on to create the maps of contamination using small desktop mapping (GIS) system. The system is powered from inner batteries or from external 12V car batteries. The system is designed to be used in measurements on foot, in a car, with helicopter or small fixed wing aircraft.



## RADIATION SITUATION IN THE CZECH REPUBLIC IN 1994

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### Abstract

The data on radiation situation obtained by the Radiation Monitoring Network of ČR are collected and evaluated by the Centre of Radiation Monitoring Network and presented as the Annual Report on the Radiation Situation on the territory of ČR. This Report is divided into two principal parts:

- monitoring for the purpose of timely detection of an not noticed nuclear accident especially abroad. This part include also monitoring of the consequences of the Chernobyl accident.
- monitoring of effluents and radiation situation in the vicinity of the NPPs.

The results from 1994 Annual Report can be summarized in the following way:

- committed effective dose equivalent from the intake of cesium radioisotopes due to Chernobyl accident was about 4  $\mu\text{Sv}$ , effective dose equivalent from the external irradiation (from contaminated surfaces) was about 5.3  $\mu\text{Sv}$
- the specific activity of cesium radioisotopes in aerosols, fallout, water, foodstuff and other environmental samples was very low (in most cases below the detection limits), the average  $^{137}\text{Cs}$  concentration in aerosols was about 2  $\mu\text{Bq}/\text{m}^3$ , the average monthly fallout of  $^{137}\text{Cs}$  was about 0.2  $\text{Bq}/\text{m}^2$ , the activity concentration of  $^{137}\text{Cs}$  in foodstuff was within the range 0.06 (milk) to 0.4  $\text{Bq}/\text{kg}$  (beef), the activity concentration of  $^{137}\text{Cs}$  in drinking water was bellow 1  $\text{mBq}/\text{l}$ ,
- the reference group of 30 people was measured on the whole body counter of Centre of Radiation Hygiene in the framework of long term study of the retention of  $^{137}\text{Cs}$  in people which started immediately after the Chernobyl accident. The results of whole body measurements are compared with the urine sample measurements from the whole CR. The average whole body retention of  $^{137}\text{Cs}$  in 1994 was about 90 Bq,
- results of the environmental radiation monitoring in the vicinity of NPPs are similar to the results obtained by the territorial monitoring,
- surveillance of effluents from the NPP Dukovany into the atmosphere and the hydrosphere demonstrated that the measured activities of the radionuclides in releases are substantially bellow the authorized limits (with the exeption of  $^3\text{H}$ ).



## ACTIVITY OF CS-137 IN RED DEER AND WILD BOAR IN SLOVAKIA

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After the Chernobyl accident the State Veterinary Service of SR launched its own program to monitor the activities of man-made radionuclides in animal husbandry products, game animals and some basic inputs to agricultural production. Even if the number of samples was considerably cut down during last years, the program continues as a part of larger monitoring scheme on hazardous chemicals, supervised by The Ministry of Agriculture of SR.

Results of monitoring the activity of radiocesium in game animals from various parts of Slovakia are presented. Samples of game flesh were collected by veterinary officials during hunting seasons 1988 - 1994. More than 80 % of samples came from following districts of Slovakia: Žiar nad Hronom(ZH), Prievidza(PD), Martin(MT), Rimavská Sobota(RS), Senica(SE), Banská Bystrica(BB), Rožňava(RV), Poprad(PP) and Spišská Nová Ves(SN). All measurements were carried out in the Laboratory of radiometry and radioecology in Nitra, using gammaspectrometric system equipped with 4 HPGe detectors. Presented results were obtained using statistical evaluation for left-censored log-normal distribution of data sets [1].

The activity of Cs-137 found in muscular tissue of red deer and of wild boar is presented in tables 1 and 2, respectively. Samples with maximal values of Cs-137 in individual years are reported in connection with their overall errors, including the two sigma error due to counting statistics and the uncertainty due to efficiency calibration.

Overall activities of Cs-137 found in red deer and wild boars in Slovakia are considerably lower, than activities reported in game animals from some parts of Northern Moravia, Southern Bohemia and Austria. While the mean activities in red deer show a decreasing tendency, mean activities of wild boar are low, but with higher occurrence of extreme values, and hence, higher variance. The observed difference could be explained by the feeding habits of wild boar: grubbing in the ground for worms, larvae, roots, etc. can lead to presence of up to 20 % of contaminated soil in their stomach [2]. At the same time wild boars often graze farmlands, where the activity of the Cs-137 in the top soil layer is reduced by ploughing and radiocesium adsorption on clay particles [3]. Fraction of farmlands in the home range of the wild boars and the time of shooting could contribute to observed variations in radiocesium activity.

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**Tab. 1** Activity of Cs-137 in the muscle tissue of red deer in Slovakia

| Hunting season | Median [Bq/kg] | Mean [Bq/kg] | Variance | Maximum value site/district   | N psc | Below MDA [%] | Mean MDA [Bq/kg] |
|----------------|----------------|--------------|----------|-------------------------------|-------|---------------|------------------|
| 1988           | 7,30           | 6,47         | 1,99     | 75.3±13.7<br>Bojnice /PD      | 472   | 6,8           | 1,75             |
| 1989           | 5,24           | 5,01         | 3,03     | 142.0±20.8<br>Turč.Teplice/MT | 720   | 4,4           | 1,16             |
| 1990           | 2,40           | 2,49         | 4,04     | 160.2±19.5<br>Nitr.Rudno /PD  | 234   | 18,4          | 1,03             |
| 1991           | 2,18           | 2,24         | 2,78     | 63.2±9.9<br>Krompachy /SN     | 216   | 11,6          | 0,95             |
| 1992           | 1,35           | 1,42         | 3,81     | 61.9±3.5<br>Nálepko /SN       | 122   | 9,0           | 0,53             |
| 1993           | 1,10           | 1,35         | 5,00     | 55.9±3.1<br>Cifer /TT         | 67    | 6,0           | 0,32             |
| 1994           | 0,95           | 1,21         | 5,86     | 27.3±2.7<br>Smolník /SN       | 46    | 6,5           | 0,37             |

N ..... total number of samples  
MDA ... Minimum Detectable Activity

**Tab. 2** Activity of Cs-137 in the muscle tissue of wild boar in Slovakia

| Hunting season | Median [Bq/kg] | Mean [Bq/kg] | Variance | Maximum value site/district | N psc | Below MDA [%] | Mean MDA [Bq/kg] |
|----------------|----------------|--------------|----------|-----------------------------|-------|---------------|------------------|
| 1987           | 2,80           | 2,99         | 6,45     | 46.6±5.1<br>Považ.Chlmec/Z  | 17    | 35,3          | 1,6              |
| 1988           | 3,65           | 3,27         | 2,85     | 43.4±9.2<br>Hrušovo /RS     | 370   | 24,9          | 1,9              |
| 1989           | 2,31           | 2,10         | 2,63     | 89.5±13.6<br>Slaská /ZH     | 228   | 19,7          | 1,1              |
| 1990           | 2,91           | 2,60         | 3,73     | 49.6±7.6<br>Dechtice /TT    | 72    | 18,1          | 1,1              |
| 1991           | 2,19           | 2,62         | 6,31     | 109.8±14.8<br>Jablonov /SN  | 33    | 9,1           | 0,9              |
| 1992           | 2,69           | 2,33         | 12,61    | 98.3±5.1<br>Svit /PP        | 37    | 5,4           | 0,4              |
| 1993           | 1,02           | 1,77         | 15,44    | 130.0±7.3<br>Brezno /BB     | 15    | 6,7           | 0,3              |
| 1994           | 1,03           | 1,35         | 12,86    | 39.8±3.8<br>Hrable /SN      | 28    | 7,1           | 0,3              |

N ..... total number of samples  
MDA ... Minimum Detectable Activity



## LONG TIME CONTAMINATION FROM PLUTONIUM

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### Introduction

Plutonium is known to be carcinogenic in animals, with tumours of bone, lung and liver representing the cancers of primary concern (Voelz 1991). The distribution of <sup>239</sup>Pu in a human whole body is reported in the research program of the United States Transuranium Registry (USTR) (Voelz et al. 1979, J.F.McInroy et al. 1991). Some cases of osteogenic sarcoma have been observed among the workers with plutonium (Voelz and Lawrence 1991) and the lung cancer, too (Wiggs et al. 1994).

Everyone can know the cases of the mortality after accident of Nuclear power plant in Chernobyl, but nothing was published about the health of the workers who have been contaminated by plutonium.

The reason, why we are busy with this problem was the request of the chief of the Nuclear Medicine Clinic in Martin to perform a control analysis of the content of the plutonium in the urine of a patient S.R.. It concerns a former citizen of the Ukraine ( year of birth 1959 ), who had participated in the liquidation works after the Chernobyl accident. For three month he had stayed in the epicentre, where he acted as a chauffeur driving a radioactive material to the place of destination.

Until the year 1986 he had never been ill, consequently he has not any health card. S.R. hasn't undergone give any health control since the finish liquidation works in the Chernobyl. For patient's repeated complaints , about headache and a loss of muscular strength in the extremities, he was examined at the clinic of the Nuclear Medicine in Charkov in the 1990. In the year 1991 he was submitted to a blood dialyse for removing radionuclides. At the present time the patient is citizen of the Slovak Republic.

### Materials and methods

We have determined plutonium in the organism of the patient from urine. To determine the concentration of <sup>239,240</sup>Pu in urine a modified radiochemical method was used. After mineralization the sample was separated as an anion-nitrate complex with contact by the anion form of the resin in the column. The resin was washed by 8 M-HNO<sub>3</sub>, then 8M-HCl-0.3M-HNO<sub>3</sub> for removing the other radionuclides. The solution 0.36M-HCl - 0.01M-HF was used for the elution of Pu. Using the lanthanum fluoride technique (Joshi 1985), the sample was filtrated through a membrane filter. The plutonium was detected in the dry sample. The <sup>239</sup>Pu tracer was used for the evaluation of the plutonium separation efficiency.

The alpha - spectrometric measurements were carried out with a large area silicon detector PD-900-100-AM Canberra. The samples were measured and evaluated in the energy region 4.98 - 5.18 MeV. The detection limit of alpha-spectrometry measurements has been 10<sup>-2</sup> Bq .l<sup>-1</sup>.

### Results and discussion

The concentration of plutonium in the 24-hour urine we determined three times in the quarter year intervals. The results are: 54mBq, 63,2 mBq, 53 mBq, with average 56,7 mBq. Because the urine of patient S.R. contained a lot of saline, the results are determined with relative large error of about 20%.

From the results of the analyses of plutonium depositions calculated according to ICRP [ICRP 54] the intake of this radionuclide for our patient was 56.7 kBq. To estimate a committed effective dose ( 50 years ) from the intake of plutonium we used a conversion factor  $6.8 \cdot 10^{-5}$  Sv.Bq<sup>-1</sup> (class W ) (IAEA 1994). So the expressed committed effective dose received from the plutonium intake is 3.8 Sv.

This number is relatively high and all the effective dose will be higher, because the patient was exposed to the other radionuclides too. For example the determination of the rate radionuclides <sup>241</sup>Am/<sup>239,240</sup>Pu was 32-36 % in the fallout after the Chernobyl accident, so the activity of <sup>241</sup>Am calculated on the basis of the plutonium activity will be 19 kBq, with the committed effective dose 1.3 Sv.

The result from the last examination of the patient at the Clinic in Martin is :

- Encephalopathy with a psychoorganic syndrome and less psychomotoric function, damage of CNS as consequence irradiation and acroparestesia of the legs
- Diabetes insipidus with a tubular damage of the kidney
- Diffusion lesion of the liver in the s. steatosis
- A subclinic form of the hypothyreosis
- Muscular hypertonicus.

The results of the biochemical tests are in normal range except of the liver test where the result is increased.

To conclude, for calculation it will be necessary to modify the effective dose, respectively the absorbent dose when we take all sources of radiation, with which the patient was contaminated.

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## Whole body counting of $^{137}\text{Cs}$ nine years after Chernobyl

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### Introduction

Nine years after Chernobyl accident the activity of  $^{137}\text{Cs}$  in inhabitants of SR is on level of about 20-30 Bq which is in the region of minimal detectable activity (MDA) of common whole body equipments. Laboratories interested in whole body counting of  $^{137}\text{Cs}$  have to improve their equipments in order to decreasing of the MDA. Ways such as accomplished detection units and/or shielding improvement of the whole body counter (WBC) are expensive.

The other method of decreasing of the MDA of the WBC is in use of information contained in the measured spectra. For illustration, pulse height spectra of HPGe detector in field of the 0.662 MeV photons and photons escaped from surface of phantom with  $^{137}\text{Cs}$  are shown in Fig.1. The spectra were calculated by Monte Carlo method.

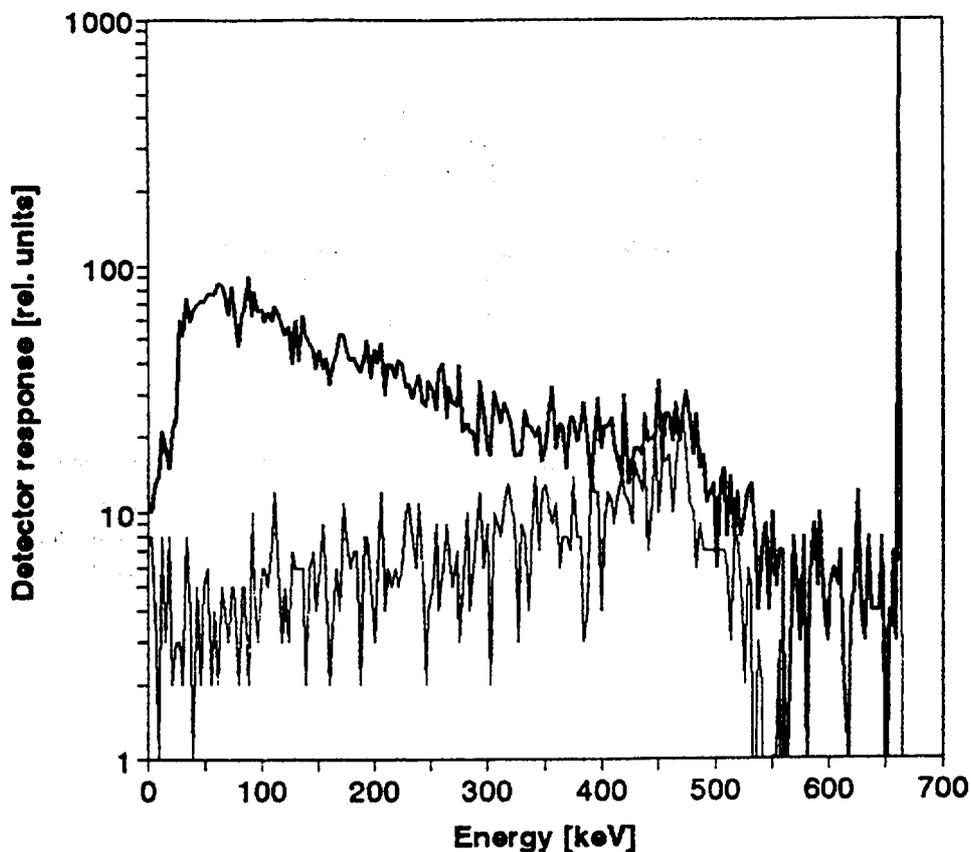
The information on Compton continuum (in Fig.1 is shown as the normal line) was used in unfolding method. The result of the unfolding method is shown in Fig.2 where the photon energy spectrum at phantom of standard men filled by set of calibration sources was measured by HPGe detector with relative efficiency 61.7% of 7.62x7.62 cm NaI detector for the  $^{60}\text{Co}$  1.33 MeV gamma-ray peak. The photon fluence of the spectrum is composed from peaks of primary photons and from continuums of photons scattered in the phantom. The radionuclide activities in the phantom are calculated by transfer coefficients of fluence of primary photons to activity in the body. By this unfolding method can be significantly decreased the value of MDA [1].

An additional improvement in the unfolding method is based on using the detector response to photons scattered in the measured body, bold line in Fig.1. The area under pulse height spectrum from all photons escaped from measured body is greater then in the previous case. Thus information from experimental spectra based on detection of the all photons emitted by phantoms are higher quality than that based on detection of the primary photons. The results of the presented unfolding method are peaks of photons emitted in the measured body without background of photons scattered in the body.

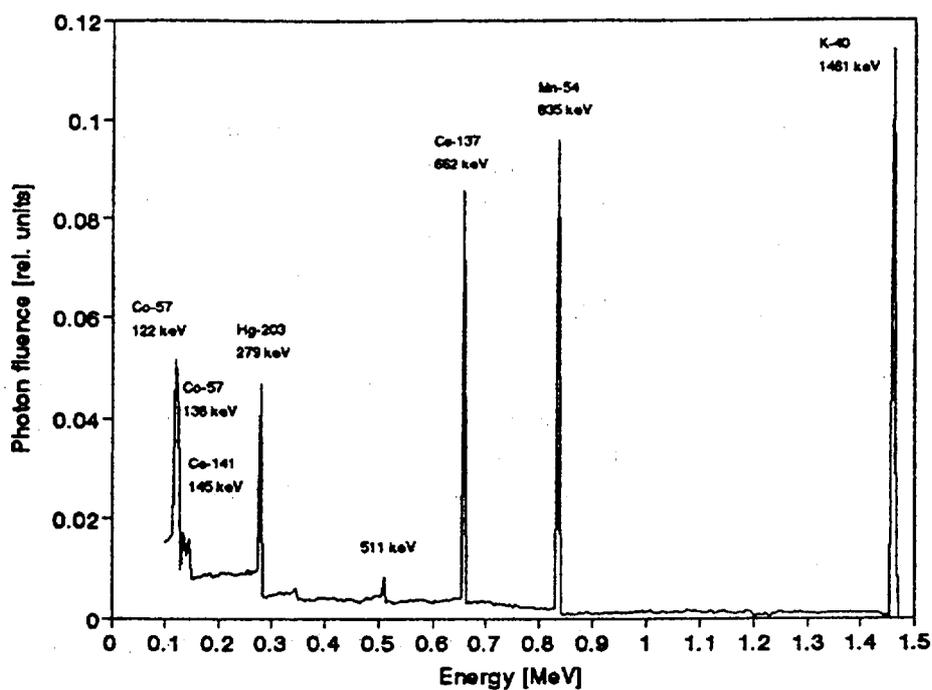
The detector responses to photons (primary and scattered, as well) escaped from phantom of standard men has been calculated by Monte Carlo method. HPGe detector (of relative efficiency 61.7%) was located at distance of 2 cm from the surface of supine phantom opposite to the small intestine. The response matrix has been calculated for homogeneously distributed  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in phantom of standard man and for Rn progeny concentrated in lungs.

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**Fig.1** Pulse height spectrum of HPGe detector (of relative efficiency 61.7%) to 0.662 photons. The spectrum at phantom surface filled by homogeneously distributed  $^{137}\text{Cs}$  source is drawn by bold line.



**Fig.2** The photon energy spectrum unfolded from pulse height spectrum measured by HPGe detector (of relative efficiency 61.7%) located at phantom of standard men filled by water solution of various calibration gamma source.



## INTERCOMPARISONS IN THE RADIATION MONITORING NETWORK OF THE CZECH REPUBLIC

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### Summary

In Czech Republic, altogether 11 laboratories, equipped by semiconductor gamma spectrometry supply regularly to the Centre of Radiation Monitoring Network the measured data about the radionuclide activity concentration in different environmental samples, participating thus in monitoring of radiation situation in the country. The Center of Radiation Monitoring Network (RMN) of Czech Republic periodically organizes through its reference laboratories interlaboratory comparison tests ensuring thus quality of the measurement within the radiation monitoring network.

A ring intercomparison test was organized in 1994. The piece of steel rather highly contaminated by <sup>60</sup>Co was used. In the intercomparison test 1994-1995, samples of pulverized concrete breeze-block containing fly-ash with natural radionuclides were used. Results of this measurement is given as an example.

Summary of reported radionuclide activity concentration in pulverized concrete breeze block

| Nuclide | Number of lab. | Arithmetic mean [Bq/kg] | Range of measure values [Bq/kg] | Confidence interval ( $\alpha = 0.05$ ) [Bq/kg] |
|---------|----------------|-------------------------|---------------------------------|---|
| K-40    | 10             | 386                     | 360-413                         | 373-399   |
| Ac-228  | 10             | 62                      | 53-78                           | 56-67   |
| U-235   | 10             | 5,5                     | 3.0-8.9                         | 4.1-6.6   |
| Ra-226  | 10             | 79                      | 69-91                           | 74-83   |
| Pb-210  | 6              | 54                      | 13-89                           | 33-75   |



## INTERNATIONAL SYSTEMS FOR HARMONIZATION AND QUALITY CONTROL BY IN VIVO MONITORING OF INTERNAL CONTAMINATION

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### **Summary**

After the Chernobyl accident, whole body counting for the monitoring of internal contamination of the population was widely used. In many cases, great discrepancy occurred between in vivo measured values and predicted ones from model calculations. This situation was very similar to the situation in environmental monitoring, however, by sample measurement, quality assurance and quality control of the measurement procedure is much easier than by whole body counting. In addition to it, unlike in environmental monitoring, use of semiconductor detectors by whole body counting was rather limited in this time so there were only few laboratories which were able to distinguish up to 20 radionuclides by which people from the vicinity of Chernobyl were internally contaminated. Therefore, effort was undertaken by many international organization to summarize current status of whole body counting and to estimate uncertainty and sensitivity of different devices.

There are efforts also to harmonize interpretation of measured results.

As a result of such activities, recommendation of international organization occurred or are under preparation. International Atomic Energy Agency issued publication Rapid monitoring of large groups of internally contaminated people following a radiation accident (IAEA-TECDOC-746, May 1994) and prepares Direct methods for measuring radionuclides in the human body (IAEA Safety Practice 147), European Commission issued Guidance notes on the calibration of whole -body counters and on the interpretation of the measured results (EUR 15395 EN, 1991), ICRU prepares report: In vivo determination of body contents of radionuclides. Also numerous intercomparison - intercalibration of the whole body counters are underway. Human Monitoring Laboratory of Radiation Protection Bureau from Ottawa, Ontario, Canada shipped in 1993-1994 BOMAB phantom of 4-year old child altogether to 35 laboratories in 4 continents. At present time, intercomparison using Reference Female phantom is prepared by HML. European Commission organizes " 1995 Intercomparison of in-vivo Monitoring System" during which phantom with unknown mixture of radionuclides will be measured in more than 40 European whole body counters. Intercomparison of calculated intakes from measurement and intercalibration using internally contaminated person is part of EC Programme. EURADOS Working Group 6: Internal Contamination prepared 9 examples using results of whole body counting and excretion analysis from real cases. As an international exercise serve calculation of intakes and doses for these cases. Whole body counting laboratory of the National Radiation Protection Institute in Prague participates in the above mentioned intercomparisons with good results and scientists from NRPI have participated in the preparation of 3 international recommendations concerning measurement of internal contamination.



## ACTIVITY OF CS-137 IN SOME FOODSTUFFS IMPORTED TO SLOVAK REPUBLIC

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Laboratory of radiometry and radioecology in Nitra was established in 1987 after the Chernobyl accident. One of its main aim is the monitoring of agricultural production as from Slovak territory as imported from other countries.

In this paper the results of Cs-137 activity in some foodstuffs imported to Slovak Republic are presented. The results are discussed from two points of view:

1. sort of foodstuffs (we chose two sort - the fish and fish products and beef, pork and bowels)
2. country of origin (we chose three countries - Germany, Norway and the Ukraine).

The above mentioned choice is based mainly on staistical grounds, we evaluated data sets with total population at least 100, the only exception is the data set for the Ukraine. For evaluation of our data sets the statistical procedures developed for left-censored data sets with assumption of lognormal distribution were used [1]. The analyzed data sets were obtained by semiconductor gamma spectrometry in Laboratory of Radiometry and Radioecology.

The results are reported in six tables with basic statistical characteristics like number of samples, number of samples with activity >MDA, median, arithmetic mean, variance and maximum value.

In tab.1 there are results for fish and fish products (concerning mainly sea fish and products). In general we can conclude that fish belong to the most contaminated sort of foodstuffs. More then 50% of samples were positively identified as Cs-137 containing.

From tab.2 (beef and pork) we can see that this sort of foodstuffs is considerable less contaminated than fish. This sort of foodstuffs is contaminated comparable to those from Slovak and Czech Republic [2,3].

In the case of countries (tab.3, tab.4 and tab.5) the most contaminated foodstuffs come from Norway, what can be explained by composition of imported foodstuffs (mainly sea fish).

In tab.6 the ten highest values from all data set (for all sorts of foodstuffs measured within 8 years - 1024 samples in all) are reported. Except one sample, beef with the highest activity ( $80.2 \pm 4.9$ ) Bq/kg from Finland, the rest are sea fish from Baltic countries, but unfortunately the origin of analyzed fishes is unknown, so we can make no decision about Baltic sea.

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**Tab.1** Activity of Cs-137 in fish and fish products

| YEAR | N   | >MDA | MEDIAN | MEAN | VAR. | MAX. VALUE          |
|------|-----|------|--------|------|------|---------------------|
| 1988 | 28  | 17   | 1,06   | 0,98 | 2,68 | 5.80 +/- 1.00 NOR.  |
| 1989 | 30  | 23   | 1,20   | 1,04 | 1,95 | 4.82 +/- 1.25 NOR.  |
| 1990 | 62  | 40   | 0,63   | 0,82 | 3,51 | 15.55 +/- 2.30 USSR |
| 1991 | 37  | 12   | 0,33   | 0,38 | 1,64 | 2.36 +/- 0.40 DENM. |
| 1992 | 60  | 34   | 0,40   | 0,71 | 6,21 | 10.65 +/- 0.96 USSR |
| 1993 | 172 | 73   | 0,34   | 0,53 | 3,41 | 12.82 +/- 1.08 EST. |
| 1994 | 103 | 71   | 0,36   | 0,49 | 2,99 | 11.73 +/- 0.89 POL. |
| 1995 | 109 | 62   | 0,26   | 0,34 | 2,98 | 11.24 +/- 0.75 LIT. |
| SUM  | 601 | 332  | 0,34   | 0,54 | 3,51 | 15.55 +/- 2.30 USSR |

**Tab.2** Activity of Cs-137 in beef, pork and bowels

| YEAR | N   | >MDA | MEDIAN | MEAN | VAR. | MAX. VALUE          |
|------|-----|------|--------|------|------|---------------------|
| 1988 | 10  | 7    | 1,73   | 1,15 | 2,93 | 3.80 +/- 0.80 GDR   |
| 1989 | 7   | 7    | 1,40   | 0,97 | 1,58 | 2.10 +/- 0.70 GDR   |
| 1990 | 10  | 5    | 0,35   | 0,40 | 1,49 | 1.07 +/- 0.34 GDR   |
| 1991 | 20  | 2    | 0,38   | 0,42 | 1,09 | 0.76 +/- 0.21 HUN.  |
| 1992 | 89  | 31   | 0,30   | 0,47 | 2,85 | 6.26 +/- 0.66 LIT.  |
| 1993 | 40  | 6    | 0,28   | 0,30 | 1,19 | 1.10 +/- 0.24 DEN.  |
| 1994 | 37  | 7    | 0,30   | 0,38 | 3,23 | 80.19 +/- 4.92 FIN. |
| 1995 | 5   | 3    | 0,21   | 0,29 | 7,26 | 3.13 +/- 0.28 GER.  |
| SUM  | 218 | 68   | 0,30   | 0,43 | 2,38 | 80.19 +/- 4.92 FIN. |

**Tab.3** Activity of Cs-137 in foodstuffs from Germany

| YEAR | N   | >MDA | MEDIAN | MEAN | VAR. | MAX. VALUE         |
|------|-----|------|--------|------|------|--------------------|
| 1988 | 11  | 8    | 0,7    | 1,04 | 2,90 | 3.80 +/- 0.80 pork |
| 1989 | 17  | 13   | 1,10   | 0,81 | 1,82 | 2.90 +/- 0.70 fish |
| 1990 | 33  | 21   | 0,60   | 0,83 | 3,50 | 8.19 +/- 1.83 fish |
| 1991 | 10  | 3    | 0,33   | 0,34 | 1,14 | 0.65 +/- 0.36 fish |
| 1992 | 13  | 4    | 0,27   | 0,28 | 1,36 | 0.72 +/- 0.22 fish |
| 1993 | 23  | 8    | 0,41   | 0,41 | 1,29 | 1.61 +/- 0.31 fish |
| 1994 | 26  | 9    | 0,25   | 0,27 | 1,57 | 2.32 +/- 0.43 fish |
| 1995 | 49  | 20   | 0,24   | 0,26 | 1,64 | 3.13 +/- 0.28 beef |
| SUM  | 182 | 86   | 0,34   | 0,42 | 2,36 | 8.19 +/- 1.83 fish |

**Tab.4** Activity of Cs-137 in foodstuffs from Norway

| YEAR | N   | >MDA | MEDIAN | MEAN | VAR. | MAX. VALUE         |
|------|-----|------|--------|------|------|--------------------|
| 1988 | 14  | 10   | 1,65   | 1,36 | 2,23 | 5.80 +/- 1.00 fish |
| 1989 | 11  | 10   | 1,50   | 1,48 | 1,70 | 4.82 +/- 1.25 fish |
| 1990 | 12  | 11   | 0,84   | 0,78 | 1,31 | 1.50 +/- 0.71 fish |
| 1993 | 27  | 19   | 0,79   | 0,72 | 1,61 | 2.12 +/- 0.40 fish |
| 1994 | 43  | 30   | 0,34   | 0,33 | 1,18 | 0.94 +/- 0.12 fish |
| 1995 | 15  | 9    | 0,41   | 0,38 | 1,37 | 1.15 +/- 0.26 beef |
| SUM  | 123 | 90   | 0,47   | 0,58 | 1,98 | 5.80 +/- 1.00 fish |

**Tab.5** Activity of Cs-137 in foodstuffs from the Ukraine

| YEAR | N  | >MDA | MEDIAN | MEAN | VAR. | MAX. VALUE          |
|------|----|------|--------|------|------|---------------------|
| 1992 | 14 | 7    | 0,29   | 0,64 | 8,57 | 6.32 +/- 0.49 eligo |
| 1993 | 3  | 3    | 0,34   | 0,45 | 1,70 | 1.03 +/- 0.22 pork  |
| 1994 | 26 | 11   | 0,30   | 0,25 | 1,63 | 0.92 +/- 0.43 eligo |
| 1995 | 17 | 4    | 0,19   | 0,19 | 1,23 | 0.36 +/- 0.30 eligo |
| SUM  | 60 | 25   | 0,25   | 0,30 | 2,68 | 6.32 +/- 0.49 eligo |

**Tab.6** The ten highest values of Cs-137 and Cs-134 activity

| FOOD | COUNTRY  | YEAR | CS-137 ACTIVITY | CS-134 ACTIVITY |
|------|----------|------|-----------------|-----------------|
| PORK | FINLAND  | 1994 | 80.19 +/- 4.92  | 3.03 +/- 0.41   |
| FISH | USSR     | 1990 | 15.55 +/- 2.30  | 1.97 +/- 0.47   |
| FISH | DENMARK  | 1990 | 13.70 +/- 2.54  | 1.64 +/- 0.60   |
| FISH | ESTONIA  | 1993 | 12.82 +/- 1.08  | 0.81 +/- 0.17   |
| FISH | ESTONIA  | 1993 | 12.12 +/- 0.80  | 0.82 +/- 0.10   |
| FISH | POLAND   | 1994 | 11.73 +/- 0.89  | 0.50 +/- 0.36   |
| FISH | LATVIA   | 1993 | 11.42 +/- 0.82  | 0.69 +/- 0.12   |
| FISH | LITHUANI | 1995 | 11.24 +/- 0.75  | 0.38 +/- 0.10   |
| FISH | POLAND   | 1993 | 11.14 +/- 0.88  | 0.70 +/- 0.13   |
| FISH | DENMARK  | 1990 | 10.70 +/- 2.20  | 1.40 +/- 0.60   |



## THE APPLICATION OF FACTOR ANALYSIS FOR WHOLE BODY GAMMA SPECTRA WORK UP

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### Introduction

Sensitivity of whole body (WB) counting of  $^{137}\text{Cs}$  with regard to average activity present in human body (20-30 Bq) is the basic problem of measurement of this isotope at present. One of the ways how to increase sensitivity is to build monstrous equipment using some tenth of scintillation crystals and deconvolution of scintillation spectra or to prolongate the time of counting by multiple repeating. By using HPGe detector the sensitivity increase is usually done by procuring the larger crystal volume or using up to four detectors. These possibilities have more disadvantages such as high cost or are time consuming.

Using scaling confirmatory factor analysis (SCFA) for WB spectra work up is suitable alternative which do not require further detectors or equipment. The SCFA provides increase of sensitivity when the peak of analyzed nuclide is laid on a compton continua from gammas of higher energies [Km93]. The WB counting of  $^{137}\text{Cs}$  is such a type of problem - peak of  $^{137}\text{Cs}$  ( $E_g=662$  keV) is loaded on the compton continuum of  $^{40}\text{K}$  ( $E_g=1461$  keV). Typical activity 3 - 4 kBq of potassium contained in human body results into a relatively high background continuum in the region of caesium peak and can not be removed by no way.

### Methods

The activity is usually determined by

$$A_r = \frac{n - n_b}{\epsilon \cdot Y} \quad (1)$$

where  $n$  and  $n_b$  are count rates (peak net area) of measured person and background, respectively, [ $\text{s}^{-1}$ ],  $\epsilon$  is the efficiency of detector for WB counting,  $Y$  the yield of analyzed gamma line.

In the case we know to determine the photon fluence rate in a place of the detection  $F(E)$ , we need transfer function  $k(E)$  between the photon fluence rate and the activity in the body of a measured person. The activity is then determined by

$$A_s = \frac{k(E) \cdot \Phi(E)}{Y} \quad (2)$$

The transfer factor  $k(E)=1/F_0(E)$  was determined elsewhere [Fül95] whereby  $F_0(E)$  is the photon fluence in the place of detection resulting from of 1 Bq activity in a calibration phantom.  $F_0(E)$  is depicted in Fig.1.

### Experimental procedures

Experimental measurements were performed in a room shielded by walls made from pre-World War II naval armour plate steel (thickness of 15 cm). The inside of the room is lined

with 3 mm of low-activity lead and 5 mm Plexiglas. The inner dimensions of the room are 1,8 x 2,5 x 2,0 m.

The HPGe coaxial detector with relative efficiency 12,5% (in comparison with 3x3" NaI scintillator at 1332 keV peak of  $^{60}\text{Co}$ ) was used in the experiment. The distance of detector to body of 3 cm was chosen. The background was measured in the same conditions with the BOMAB type phantom of an adult man filled with distilled water.

### Results and discussion

The spectra of two person marked as WB spectrum 1 (WB1), WB spectrum 2 (WB2) and the background (bg) have been measured and analyzed. They are pictured in the 256 channel distribution, gained by summing of the 4k spectra, in Fig.2. The energy region around the  $^{137}\text{Cs}$  662 keV peak of the 4k spectra is shown on Figs.3a and 3b together with measuring times.

The spectra of photon fluence rate obtained by work up of the measurements by SCFA technique [Krn95] in the energy region 500 - 740 keV as well as in the region up to 1800 keV are on Figs.4 and 5 respectively.

The photon fluence rates obtained from the measured spectra (v256f), from the measured spectra after the background subtraction (v256fb) and pure background photon fluence rates (bg) for nuclides  $^{137}\text{Cs}$  and  $^{40}\text{K}$  are in Tables 1a and 1b respectively.

The activities  $A_k$  and  $A_\phi$ , we are interested in, have been determined by the equations (1) and (2), respectively. The results of the calculations are in Tables 2a and 2b. For the values of photon fluence rates in the equation (2) average given by  $((v256f-bg)+v256fb)/2$  has been used.

### Conclusion

The results of WB counting with small HPGe detector were presented. The SCFA method based on factorisation of the response operator is very sensitive and for this application suitable method how to decrease limits of detection. The Minimal Detectable Activity (MDA, for counting time of person 7200 s, background 58600 s and 99% confidence level) of detector usually used in our laboratory for WB counting (rel. eff. 61,8%) 18,5 Bq and MDA for the SCFA method for small detector 17,9 Bq are very close. The use of SCFA method improves the sensitivity (MDA) by factor of 4,1 and the small detector is comparable in sensitivity with the larger one.

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Tab. 1a

| Energy [keV] | WB spectrum 1         |                       | Background (bg)      | v256-bg               | Difference [(v256f-v256fb)-bg] |
|--------------|-----------------------|-----------------------|----------------------|-----------------------|--------------------------------|
|              | v256f                 | v256fb                |                      |                       |                                |
| 660          | $18,1 \cdot 10^{-4}$  | $9,9 \cdot 10^{-4}$   | $7,3 \cdot 10^{-4}$  | $10,8 \cdot 10^{-4}$  | $0,9 \cdot 10^{-4}$            |
| 1 460        | $175,6 \cdot 10^{-4}$ | $162,0 \cdot 10^{-4}$ | $12,9 \cdot 10^{-4}$ | $162,7 \cdot 10^{-4}$ | $0,7 \cdot 10^{-4}$            |

**Tab. 1b**

| Energy [keV] | WB spectrum 2         |                       | Background (bg)      | v256f-bg              | Difference [(v256f-v256fb)-bg] |
|--------------|-----------------------|-----------------------|----------------------|-----------------------|--------------------------------|
|              | v256f                 | v256fb                |                      |                       |                                |
| 660          | $14,3 \cdot 10^{-4}$  | $5,1 \cdot 10^{-4}$   | $7,3 \cdot 10^{-4}$  | $7,0 \cdot 10^{-4}$   | $1,9 \cdot 10^{-4}$            |
| 1 460        | $154,9 \cdot 10^{-4}$ | $140,6 \cdot 10^{-4}$ | $12,9 \cdot 10^{-4}$ | $142,0 \cdot 10^{-4}$ | $1,4 \cdot 10^{-4}$            |

**Tab. 2a**

|            | $n_{660}$ [s <sup>-1</sup> ]           | $A_{e660}$ [Bq]      | $\Phi_{660}$ [cm <sup>-2</sup> .s <sup>-1</sup> ] | $A_{\phi660}$ [Bq]      |
|------------|--|----------------------|---|-------------------------|
| WB spec. 1 | $(2,3 \pm 0,7) \cdot 10^{-3} \ddagger$ | $28 \pm 10 \ddagger$ | $(10,35 \pm 0,66) \cdot 10^{-4}$                  | $32,4 \pm 2,1$          |
| WB spec. 2 | —                                      | —                    | $(6,05 \pm 1,06) \cdot 10^{-4} \ddagger$          | $18,9 \pm 3,3 \ddagger$ |

$$n_{660} = n - n_b$$

‡ Values are under MDA (73 Bq) but around MSA for 99% confidence level.

† Values are under MDA (23.5 Bq).

**Tab. 2b**

|            | $n_{1460}$ [s <sup>-1</sup> ]  | $A_{e1460}$ [Bq] | $\Phi_{1460}$ [cm <sup>-2</sup> .s <sup>-1</sup> ] | $A_{\phi1460}$ [Bq] |
|------------|--------------------------------|------------------|--|---------------------|
| WB spec. 1 | $(1,9 \pm 0,17) \cdot 10^{-2}$ | $3170 \pm 280$   | $(162 \pm 5) \cdot 10^{-4}$                        | $3260 \pm 86$       |
| WB spec. 2 | $(1,9 \pm 0,23) \cdot 10^{-2}$ | $3170 \pm 380$   | $(141 \pm 7) \cdot 10^{-4}$                        | $2830 \pm 145$      |

$$n_{1460} = n - n_b$$

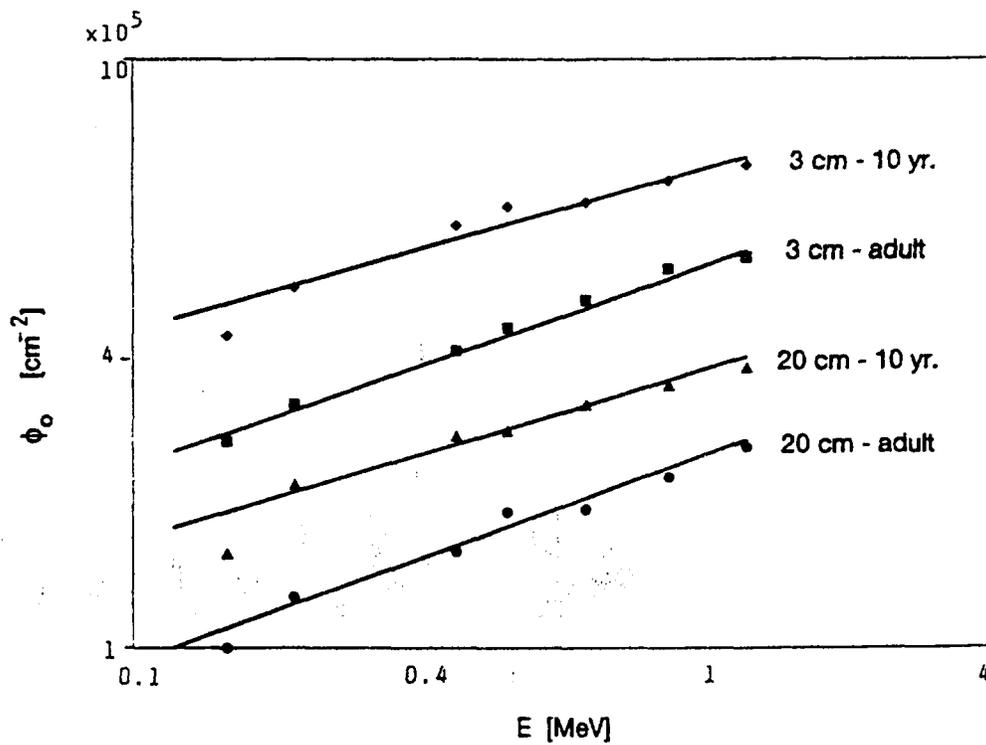


Fig.1 The integral photon fluence  $\Phi_0(E)$  for different detector-phantom distance and for 10 year old child and adult man.[Fül95]

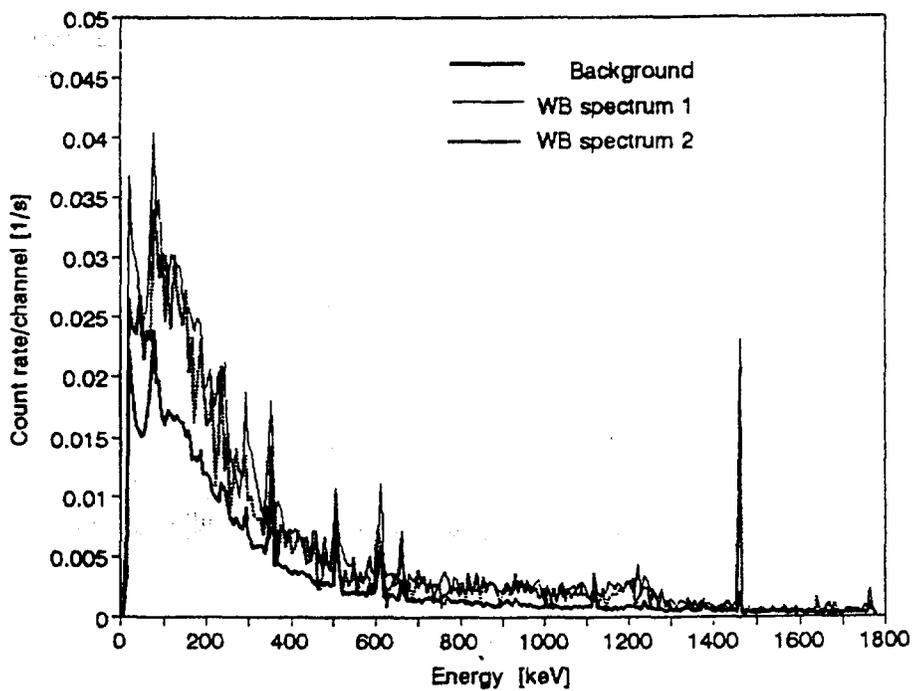


Fig.2 The measured whole body spectra in 256 channel distribution.

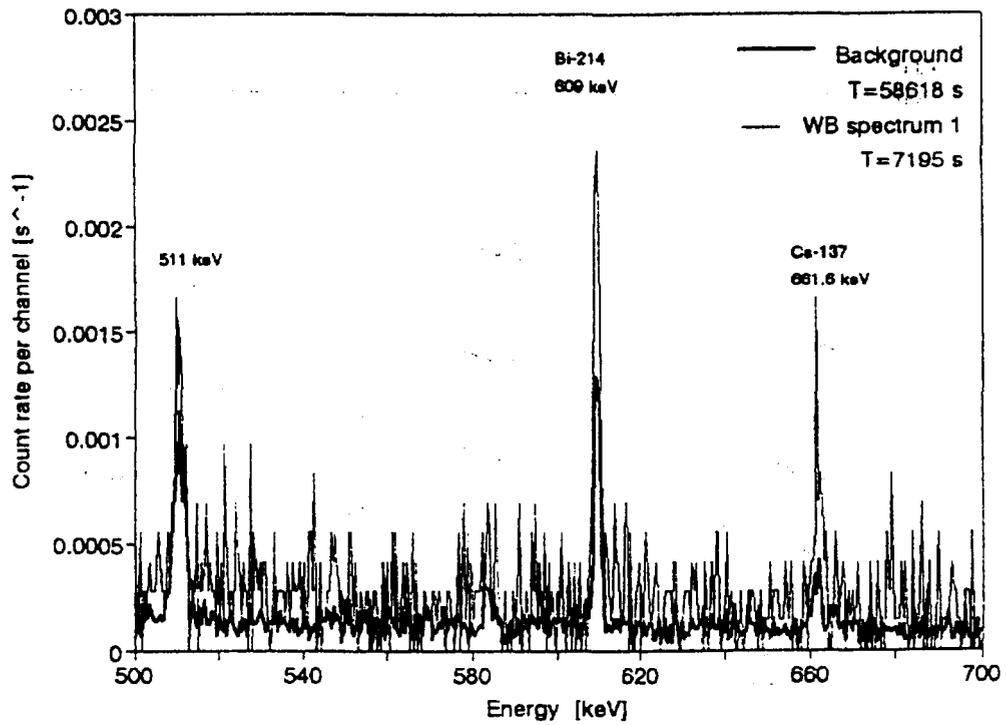


Fig.3a

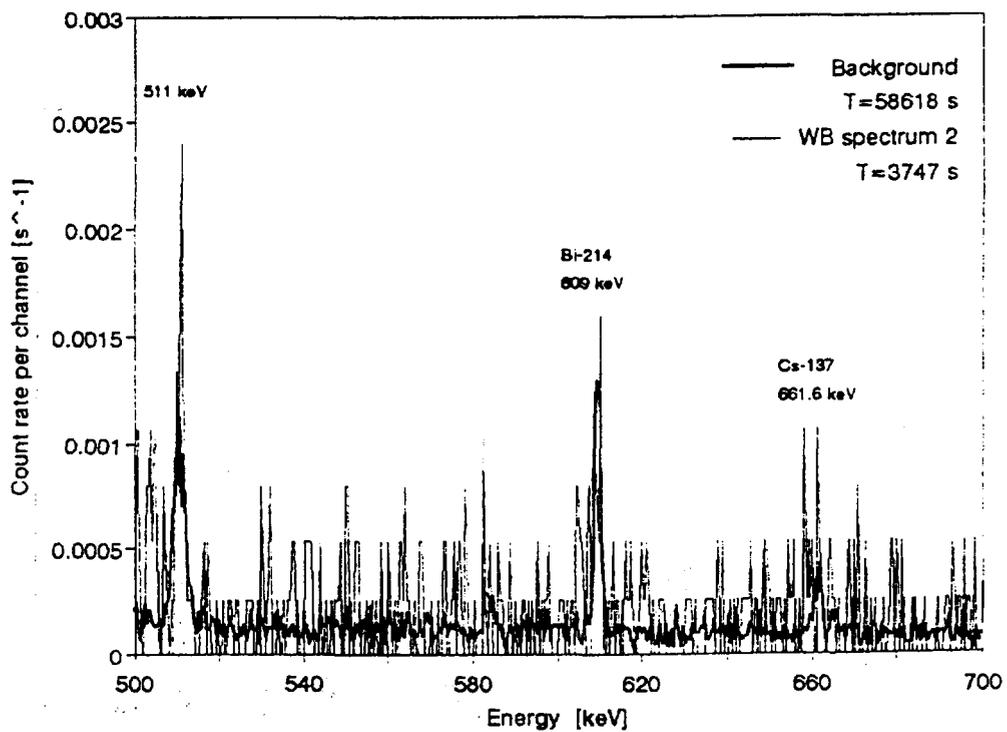


Fig.3b

Figs.3a and 3b The 500-700 keV region of the 4k channel measured spectra.

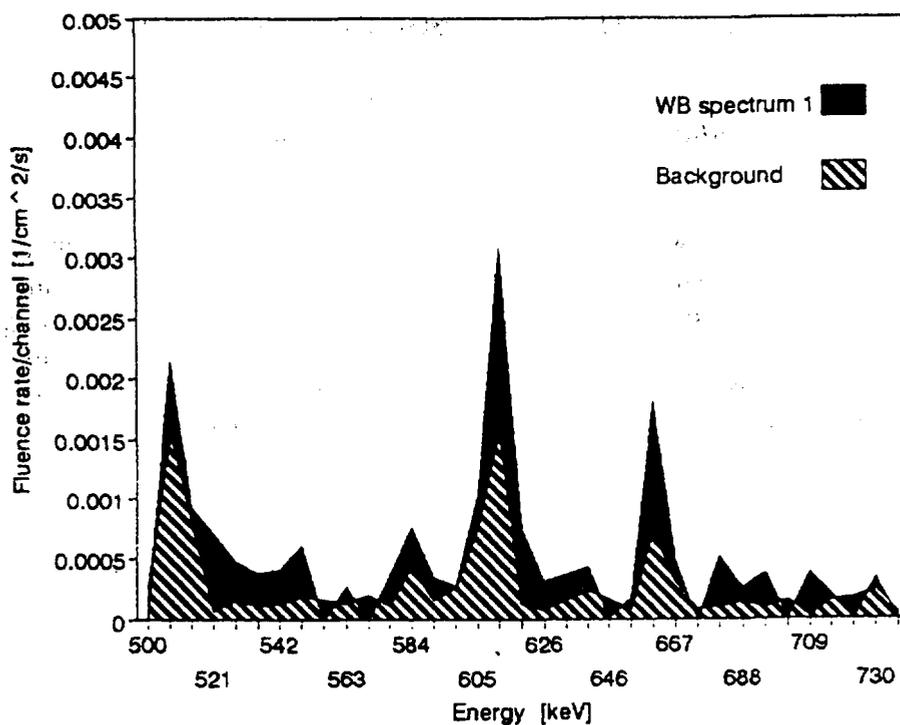


Fig.4 The spectra of photon fluence rates in energy region 500-740 keV. The background was not subtracted from WB spectra in advance.

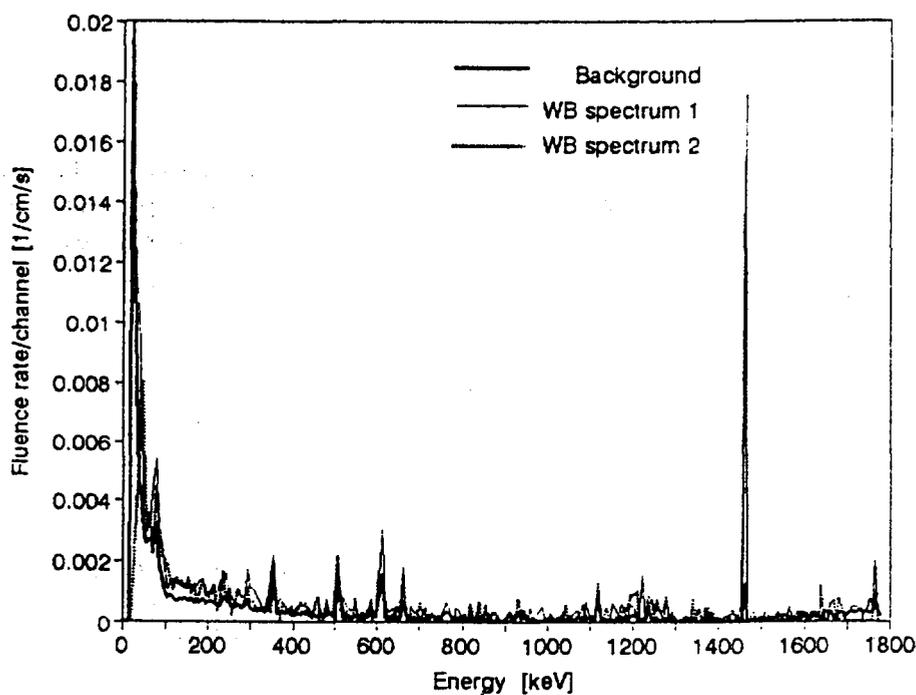


Fig.5 The spectra of photon fluence rates vs. energy. The background was not subtracted from WB spectra in advance.



## TLD TERRITORIAL NETWORK IN THE CZECH REPUBLIC

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The territorial network of thermoluminescence dosimeters is one of the most important components of the Czech Radiation Monitoring Network (CRMN). It was started in 1989 to measure the photon dose equivalent  $H_x$  in the environment on the republic's territory. Most of the measuring points have been placed outdoors where the dosimeters are exposed in free space geometry; however, some of them have also been installed in the adjoining buildings with an intention to obtain information about the shielding effectivity of inhabitants in case of a radiation accident. In case of a normal radiation situation a three months' monitoring period is used.

At present, there are 236 measuring points practically homogeneously distributed on the territory of the Czech Republic. 78 measuring points are distributed as local TLD-networks in the surroundings of our two nuclear power plants. Both of the TLD-networks are operated by the National Radiation Protection Institute in Prague.

The dosimeters hitherto used are the  $\text{CaSO}_4$ :Dy-teflon detectors placed in TL-badges and shielded on both sides by energy compensation filters composed of lead and tin.

Table 1 summarizes the results measured in the last three years in parts of the Czech Republic.

Recently, a modernization of the measuring system, type of dosimeter and methods of evaluation is under way. From 1996 the CRMN will be using the TLD system HARSHAW 4000, along with the type 8807 ENVIRONMENTAL DOSIMETER. The dosimeter is composed of four TL-elements - two LiF and two  $\text{CaF}_2$ , placed in a plastic badge. In addition, the  $\text{CaF}_2$  elements are shielded on both sides by energy compensation filters composed of tantalum and lead. Before the routine use of the new system a testing of the dosimeters was made. The following text summarizes the obtained results.

### **Results of Testing**

#### *1. Reproducibility*

Five dosimeters were exposed to 0.2 mSv of  $^{137}\text{Cs}$  ten times each. The relative standard deviation of  $\text{CaF}_2$  and LiF were found to be 6.9% and 19.1%, respectively. The substantial reproducibility difference between the LiF and the  $\text{CaF}_2$  is connected with their sensitivity difference. The  $\text{CaF}_2$  material has higher sensitivity than the LiF elements (by a factor of 20).

#### *2. Fading*

A group of dosimeters was exposed to 10 mSv of  $^{137}\text{Cs}$ . The dosimeters were stored under laboratory conditions at 25 °C. Table 2 presents the fading of LiF and  $\text{CaF}_2$  against storage period related to 1 day storage.

#### *3. Energy dependence*

The energy dependence was studied from 21 keV effective to 1250 keV effective by using standardized X and gamma radiosources. The TLD elements were shielded as described above. Data from both the LiF and  $\text{CaF}_2$  elements (normalized to  $^{60}\text{Co}$ ) are presented in Table 3.

#### 4. Linearity

Linearity was verified with  $^{137}\text{Cs}$  gammas over the range 0.2 mSv to 50 mSv, and was found to be excellently following the linear form for both materials. All the data measured are presented in Table 4.

#### Conclusion

For environmental monitoring purposes the presented results of tests of the new TLD system represent a consistent set of data on which recommendations concerning the introduction of the TLD system into routine mode can be based. It is possible also to refer good agreement of the obtained results with corresponding data of the 8807 dosimeter producer (Harshaw/Bicron - User's Manual - Environmental Dosimeter).

**Tab. 1** Average annual values (avg.) and their standard deviations (s.d.) of photon dose equivalent rate  $H_x$  [nSv/h] on the territory of the Czech Republic

| PART            | Number of measuring points |      | 1992 | 1993 | 1994 |
|-----------------|----------------------------|------|------|------|------|
| Prague          | 15                         | avg. | 99   | 89   | 96   |
|                 |                            | s.d. | 14   | 18   | 15   |
| Central Bohemia | 25                         | avg. | 113  | 105  | 106  |
|                 |                            | s.d. | 34   | 35   | 35   |
| North Bohemia   | 21                         | avg. | 108  | 109  | 114  |
|                 |                            | s.d. | 38   | 25   | 25   |
| West Bohemia    | 24                         | avg. | 115  | 116  | 132  |
|                 |                            | s.d. | 22   | 21   | 22   |
| South Bohemia   | 30                         | avg. | 120  | 110  | 119  |
|                 |                            | s.d. | 22   | 22   | 22   |
| East Bohemia    | 21                         | avg. | 99   | 94   | 97   |
|                 |                            | s.d. | 20   | 19   | 21   |
| North Moravia   | 21                         | avg. | 134  | 144  | 133  |
|                 |                            | s.d. | 26   | 26   | 20   |
| South Moravia   | 26                         | avg. | 121  | 120  | 125  |
|                 |                            | s.d. | 18   | 19   | 19   |

**Table 2 : FADING**

| Storage Period<br>[Days] | Relative Response |        |
|--------------------------|-------------------|--------|
|                          | LiF               | CaF2   |
| 1                        | 100.0%            | 100.0% |
| 3                        | 99.0%             | 87.9%  |
| 8                        | 98.6%             | 84.0%  |
| 15                       | 96.2%             | 79.9%  |
| 23                       | 90.9%             | 76.7%  |
| 42                       | 95.5%             | 75.4%  |
| 77                       | 93.7%             | 74.7%  |
| 112                      | 86.9%             | 64.9%  |

**Table 3 : ENERGY DEPENDENCE**

| E<br>[keV] | Relative Response |      |
|------------|-------------------|------|
|            | LiF               | CaF2 |
| 21         | 1.50              | 0.15 |
| 39         | 2.15              | 1.22 |
| 78         | 2.05              | 2.44 |
| 136        | 1.72              | 2.08 |
| 662        | 1.07              | 1.18 |
| 1250       | 1.00              | 1.00 |

**Table 4 : LINEARITY**

| Hx<br>[mSv] | Net Light Output (Reader Units) |         |
|-------------|---------------------------------|---------|
|             | LiF                             | CaF2    |
| 0.2         | 2.05                            | 37.95   |
| 0.4         | 3.87                            | 72.52   |
| 0.6         | 5.34                            | 104.91  |
| 0.8         | 7.10                            | 134.81  |
| 1.0         | 8.16                            | 157.75  |
| 1.4         | 11.98                           | 231.64  |
| 2.0         | 15.51                           | 298.59  |
| 4.0         | 29.81                           | 652.12  |
| 10.0        | 79.98                           | 1729.91 |
| 30.0        | 260.65                          | 5170.68 |
| 50.0        | 376.72                          | 7995.77 |



## MONITORING OF NATURAL RADIOACTIVITY IN SLOVAKIA

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### Introduction.

The Commission of the European Communities (CEC) published recently a compilation of the level of natural radiation measurement in Member States of CEC, in a form of Radiation Atlas [1]. The objective of our work was to supplement the surveyed data with an information about radiation load of Slovak population from natural sources of ionizing radiation. We review the results compiled in the years 1993 and 1994.

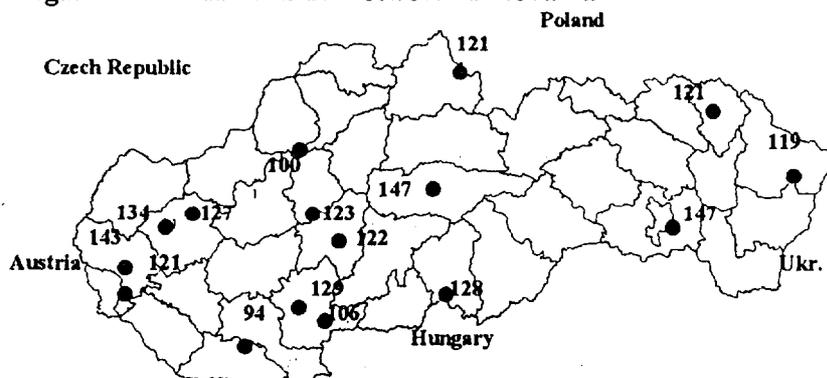
### Methods.

The paper deals with two main natural sources of human exposure, i.e. the outdoor photon dose equivalent rate, Hx, and the indoor radon volume activity [ $\text{Bq/m}^3$ ]. The estimation of the radiation load from outdoor photon radiation was based on continuous measurements of dose equivalent rates using:

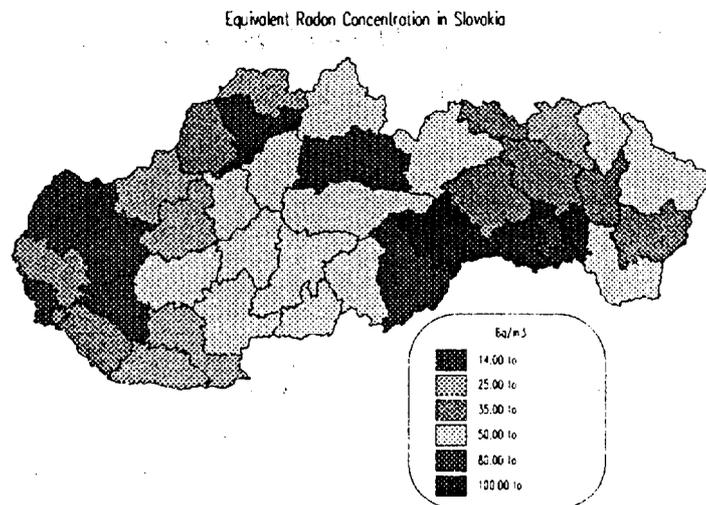
a/ TLD. types TLD 700 (LiF, Harshaw) in a territorial monitoring network, consisting of 66 stations over Slovakia, where integral values are collected for 3 months.

b/ Intelligent Environmental Dose Equivalent Proportional Detector, FHZ621B, [2] in an International Radiation Information System (IRIS). The system consists of 25 stations (16 working) over the whole Slovakia, situated uniformly at existing hydrometeorological stations (Fig.1). In standard operation a set of average dose equivalent rate, Hx, are transferred every 24 hours into the central database of IRIS [3].

Fig.1 Hx in IRIS Network in Slovakia



For the measurements of equilibrium equivalent radon concentration (EEC) solid state track detectors type CR-39 (Pershore, UK) were used, exchanged in a period of 6 month during the heating season. The survey of the distribution of EEC of radon in Slovakia is given in Fig.2.



The survey covers random sample of EEC measurements in 1832 dwellings uniformly located over the Slovakia [4].

The annual effective doses, E, from external exposure were estimated by the procedure given in UNSCEAR 1993 [5]. Conversion coefficients for radon and radon progeny were taken from Basic Safety Standards [6], UNSCEAR 93, and CEC, assuming an indoor radon occupancy of 7000 hours/year and an equilibrium factor of 0.4.

## Results.

The values of H<sub>x</sub> estimated from TLD and IRIS network are given in Tab.1. The obtained results were compared with older data of air kerma rate, K<sub>a</sub>, measured in 139 localities of Slovakia using combination of 6 area GM tubes type STS6, by Spurný at all [7].

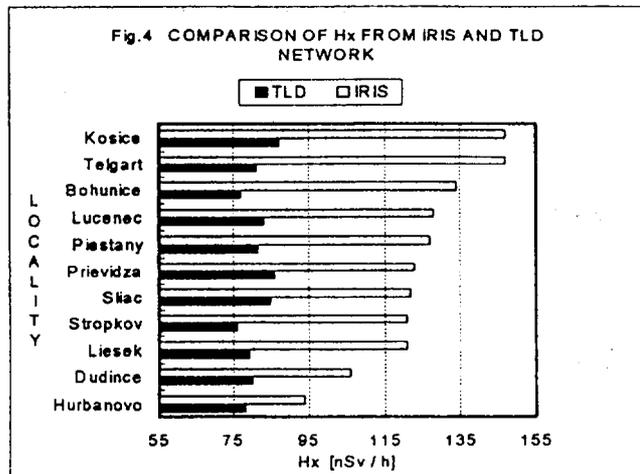
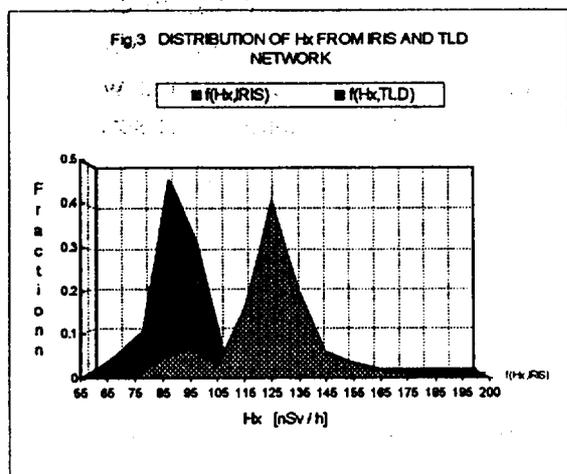
The average H<sub>x</sub> value from the IRIS network is 124 nSv/hour and from the TLD network only 95 nSv/hour. The annual effective dose is equal to 811 μSv and 618 μSv, resp.

Tab.1 ESTIMATION OF H<sub>x</sub> IN SLOVAKIA

| SOURCES      | H <sub>x</sub>          | K <sub>a</sub>          | E         | USED<br>DEVICE   |
|--------------|-------------------------|-------------------------|-----------|------------------|
|              | [nSv .h <sup>-1</sup> ] | [nGy .h <sup>-1</sup> ] | [mSv / a] |                  |
| Spurný(1977) | 124.8                   | 108.4                   | 816       | GM Tube STS 6    |
| IRIS(1994)   | 124.0                   | 811.2                   | 811       | FAG 621 B        |
| TLD (1994)   | 95.0                    | 82.5                    | 618       | <sup>7</sup> LiF |

The H<sub>x</sub> distribution from both networks are given in Fig.3. Fig.4 shows the differences of H<sub>x</sub> measured by TLD and IRIS, in the individual regions of Slovakia. At present we are investigating the reasons of discrepancy between the results of two monitoring systems used. In

this connection the energy dependence of detectors, influence of scattered radiation and the influence of buildings located near the measuring points, will be followed.



The Tab.2 gives the EEC of radon in different regions of Slovakia, together with the highest measured values. The geometric mean of EEC from 1832 dwellings is about 40 Bq/m<sup>3</sup> (it is 100 Bq/m<sup>3</sup> radon gas concentration).

Tab.2 EEC OF RADON IN DIFFERENT REGIONS SR

| REGION     | NUMBER OF HOUSES | GEOM. MEAN            | MAX. VALUE            |
|------------|------------------|-----------------------|-----------------------|
|            |                  | [Bq.m <sup>-3</sup> ] | [Bq.m <sup>-3</sup> ] |
| Slovakia   | 1832             | 40                    | 1493                  |
| Bratislava | 244              | 10                    | 906                   |
| W.Slovakia | 537              | 29                    | 1042                  |
| M.Slovakia | 583              | 58                    | 1213                  |
| E.Slovakia | 468              | 65                    | 1489                  |

The effective dose caused by the annual radon concentration of 100 Bq/m<sup>3</sup> is given in Tab.3. The values of E ranges from 1.7 to 5.0 mSv/year, depending on the used calculation procedure and published conversion factors. The mean radon concentration may be probably overestimated due to the fact, that the measured dwellings were not always strictly selected randomly.

Tab.3 COMPARISON OF EFFECTIVE DOSE FROM RADON IN DWELLINGS

| a <sub>v</sub> <sup>a)</sup> | EDCF  | E                      | Ref.         |
|------------------------------|---|------------------------|--------------|
| [Bq.m <sup>-3</sup> ]        | [μSv.a <sup>-1</sup> / Bq.m <sup>-3</sup> ] | [μSv.a <sup>-1</sup> ] |              |
| 100                          | 17.1  | 1710                   | BSS (94)     |
| 100                          | 26.8  | 2680                   | UNSCEAR (93) |
| 100                          | 50.0  | 5000                   | CEC (93)     |

<sup>a)</sup> geometric mean of radon gas in SR (Indoor)

## Conclusions.

Annual values of the effective doses from above given natural sources of radiation in Slovakia range from 1.2 mSv to 3.2 mSv/year. This broad range is caused mainly by uncertainties in calculation procedures of radon effective doses. The effective dose due to exposure to radon calculated taking into account, that an annual radon gas concentration of 20 Bq/m<sup>3</sup> is 1 mSv seems to be a very overestimated value. Therefore for protective measures we use the lower value of the above given range. This calculation methods are actually under scrutiny.

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## ON RESPONSE OPERATOR IN SEMICONDUCTOR GAMMA RAY SPECTROMETRY

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### Introduction

HPGe detectors having superior resolution are the best detectors available at present for low-level  $\gamma$ -ray spectrometry. However, their relatively lower efficiency in the MeV range resulting from technological difficulties in the production of detectors of large volumes, greatly limits the counting of ultra low activity samples (e.g. in studies of rare nuclear decays, environmental radioactivity investigations, etc. [1]).

Generally, in semiconductor  $\gamma$ -ray spectrometry [2], the most commonly used technique for evaluation of  $\gamma$ -ray spectra is the net area calculation of the full energy peak. The Peak Net Area (PNA) method provides greatly improved energy resolution, however, it takes into account only a fraction of the measured  $\gamma$ -ray spectrum.

The response operator method, mostly used in scintillation of  $\gamma$ -ray spectrometry [3], may be employed to obtain the real incident spectra. However, it is not possible to obtain an exact analytical formulation of the response function for various photon energies [3]. The application of the response operator to semiconductor  $\gamma$ -ray spectrometry is more complicated because of its higher resolution and, therefore, the greater number of spectrometric channels to be handled.

### Response operator

The results of  $c$ -channel spectra measurements, using a matrix approximation of spectra [3], may be written by  $c$ -vectors put as columns in the matrix,  $\mathbf{D}$ . Similarly, the incident  $\gamma$ -ray spectra associated with the physical spectra in  $\mathbf{D}$  are represented by  $c$ -vectors put as columns into the matrix,  $\mathbf{Q}$ .

A transformation operator converting the physical spectra (from the matrix  $\mathbf{D}$ ) into the unknown incident spectra (in the matrix  $\mathbf{Q}$ ) may be interpreted as the  $c \times c$  response matrix,  $\mathbf{K}_c$ . Therefore, the transformation is described as follows

$$\mathbf{D} = \mathbf{K}_c \mathbf{Q}. \quad (1)$$

The matrix  $\mathbf{K}_c$  represents the response operator of the measuring device and it can be estimated by measuring of point standard sources which consist of  $n$  energy components. These calibration measurements can be described by the following matrix equation

$$\mathbf{S} = \mathbf{K} \mathbf{G}, \quad (2)$$

where  $c$ -vectors as columns in the matrix,  $\mathbf{S}$ , represent the standard physical spectra and  $n$ -vectors as columns in the matrix,  $\mathbf{G}$ , represent the incident standard quantities (e.g. standard activities or photon fluence rates) corresponding to  $n$  energy components in the calibration spectra. The standard response matrix,  $\mathbf{K}$ , is the  $c \times n$  matrix of the single-energy component spectra as columns.

### Complex analysis (SCFA model)

Because suitable single-energy standard sources, required for the calibration measurements (2), are not evidently available, two important questions in connection with a complex analysis application have to be drawn: how to obtain the single-energy components in operator  $\mathbf{K}$  and how to interpolate the standard response operator,  $\mathbf{K}$ , to the complex response operator,  $\mathbf{K}_c$ . A decomposition of the response operator into a set of latent determinants (factors) uniform for all energies of the  $\gamma$ -ray spectra in the investigated region could be a good answer. This procedure is described in detail elsewhere [4].

The latent structure of the response operator is based on a set of common factors resulting from fundamental photon interaction with matter as follows:

- a) photoelectric factor,
- b) Compton factor,
- c) backscattering factor,
- d) residual factor.

Furthermore, the escape peaks appearing in the spectra at higher energies, and the other factors (e.g. the annihilation factor) should be involved.

Scaling of the common factors distributions in the matrix  $\mathbf{F}$  into corresponding fractional region in physical  $\gamma$ -ray spectra can be formulated as a two-level scaling product defined in Ref. [4] introducing the matrix of the scaling coefficients,  $\mathbf{C}$ . Then the response operator  $\mathbf{K}$  is reproduced by loading of scaling factors using weighting coefficients arranged in the matrix of factor loadings,  $\mathbf{B}$ .

The latent structure of the response operator, given by  $\mathbf{K}(\mathbf{B}, \mathbf{C}, \mathbf{F})$ , is very advantageous to formulate as a manifest covariance structure. This formalism enables us to employ many advanced computational procedures traditionally used in the confirmatory factor analysis [5]. Therefore, this method is called Scaling Confirmatory Factor Analysis (SCFA).

The latent loading, and scaling, coefficients in columns of matrix  $\mathbf{C}$  and  $\mathbf{B}$ , respectively, can be interpolated within measured energy range for energies which correspond to middles of  $c$  spectrometric channels. The complex  $c \times c$  response matrix  $\mathbf{K}_c(\mathbf{B}_c, \mathbf{C}_c, \mathbf{F})$  can be built up by the reproduction scheme (scaling and loading of the common factors) using the complete matrices,  $\mathbf{B}_c$  and  $\mathbf{C}_c$ , instead of the incomplete standard matrices,  $\mathbf{B}$  and  $\mathbf{C}$  (see Figure 1).

In the second step, complex incident  $\gamma$ -ray spectra,  $\mathbf{Q}$ , by an application of the complex response operator to unknown experimental spectra,  $\mathbf{D}$ , are obtained. As an aspect of computational performance, it has been found that an application of the maximum likelihood followed by the least square method seems to be the best compromise for a complex  $\gamma$ -ray analysis [4].

### Verification of the model

A comparison of the experimental  $\gamma$ -ray spectra and the SCFA spectra reproduced by the 4-factor hypothesis were used for a verification of the SCFA model. The verification was carried out for widely-ranged energy interval of standard point  $g$ -sources. An estimation of statistical and systematic relative errors of the model is listed in Table 1.

### Comparison with traditional approach

A comparison of sensitivity of the traditional PNA method, the integral method and the latent SCFA method based on minimum significant activity (MSA) [6] calculations are shown in Table 2. The critical level set is related to the energy at 661.6 keV ( $^{137}\text{Cs}$ ) for a point source at 10 cm from the detector forehead by using a 12.5% HPGe detector placed in a shield of 15 cm of iron.

It has been found that the sensitivity of  $^{137}\text{Cs}$  measurement for the PNA and SCFA method reaches approximately the same level in the absence of any interference. If some

interference being occurred in the spectrum, the MSA for the SCFA model is 4 to 10 times lower (depending on interferences) than for the PNA model (see Table 2).

**Tab. 1** The total, statistical and systematic errors of the SCFA model factorization.

| Nuclide           | $t_s$ [s] | $\Sigma s_i$ | $\Sigma(s_i - \xi_i)$ | $\chi^2_s$ | $\delta_{total}$ [%] | $\delta_{stat}$ [%] | $\delta_{syst}$ [%] |
|-------------------|-----------|--------------|-----------------------|------------|----------------------|---------------------|---------------------|
| $^{241}\text{Am}$ | 503       | 763,2        | 23,93                 | 15,77      | 0,65                 | 0,16                | 0,62                |
| $^{57}\text{Co}$  | 25 643    | 7,66         | 0,01                  | 31,13      | 1,26                 | 0,23                | 1,24                |
| $^{22}\text{Na}$  | 3 008     | 59,82        | 0,04                  | 1,86       | 0,32                 | 0,24                | 0,22                |
| $^{137}\text{Cs}$ | 1 041     | 2 203,4      | 12,83                 | 6,06       | 0,16                 | 0,07                | 0,15                |
| $^{88}\text{Y}$   | 3 004     | 190,56       | 0,13                  | 2,07       | 0,19                 | 0,13                | 0,14                |
| $^{60}\text{Co}$  | 1 190     | 5 851,8      | 37,61                 | 7,64       | 0,11                 | 0,04                | 0,1                 |
| Backg round       | 294 350   | 0,9          | $3,09 \cdot 10^6$     | 1,01       | 0,2                  | 0,19                | 0,02                |
| Matrix S          | 1 000     | 1 296,7      | 10,65                 | 8,21       | 0,25                 | 0,09                | 0,24                |
| $^{152}\text{Eu}$ | 1 087     | 2 920,7      | 307,6                 | 114,5      | 0,6                  | 0,06                | 0,6                 |
| <200 keV          | 1 087     | 1 388,3      | 306,8                 | 240,2      | 1,26                 | 0,08                | 1,26                |
| >200 keV          | 1 087     | 1 532,4      | 1,57                  | 1,11       | 0,08                 | 0,08                | 0,03                |

### Conclusion

Some results of the SCFA application in semiconductor  $\gamma$ -ray spectrometry presented in this contribution points out to a new ground for evaluation the  $\gamma$ -ray spectra. This whole-spectrum processing approach considerably increases detection sensitivity, especially, if significant interferences being present in the measured spectrum. Precision of the SCFA method is determined by choice of a sufficient number of suitable calibration  $\gamma$ -ray sources in the energy region of interest, by setting up an acceptable latent hypothesis and by chosen experimental quantification of spectra.

**Tab. 2.** A comparison of MSA ( $^{137}\text{Cs}$ ) for the Integral, PNA and SCFA methods (99% confidence level, counting time of 5000 sec).

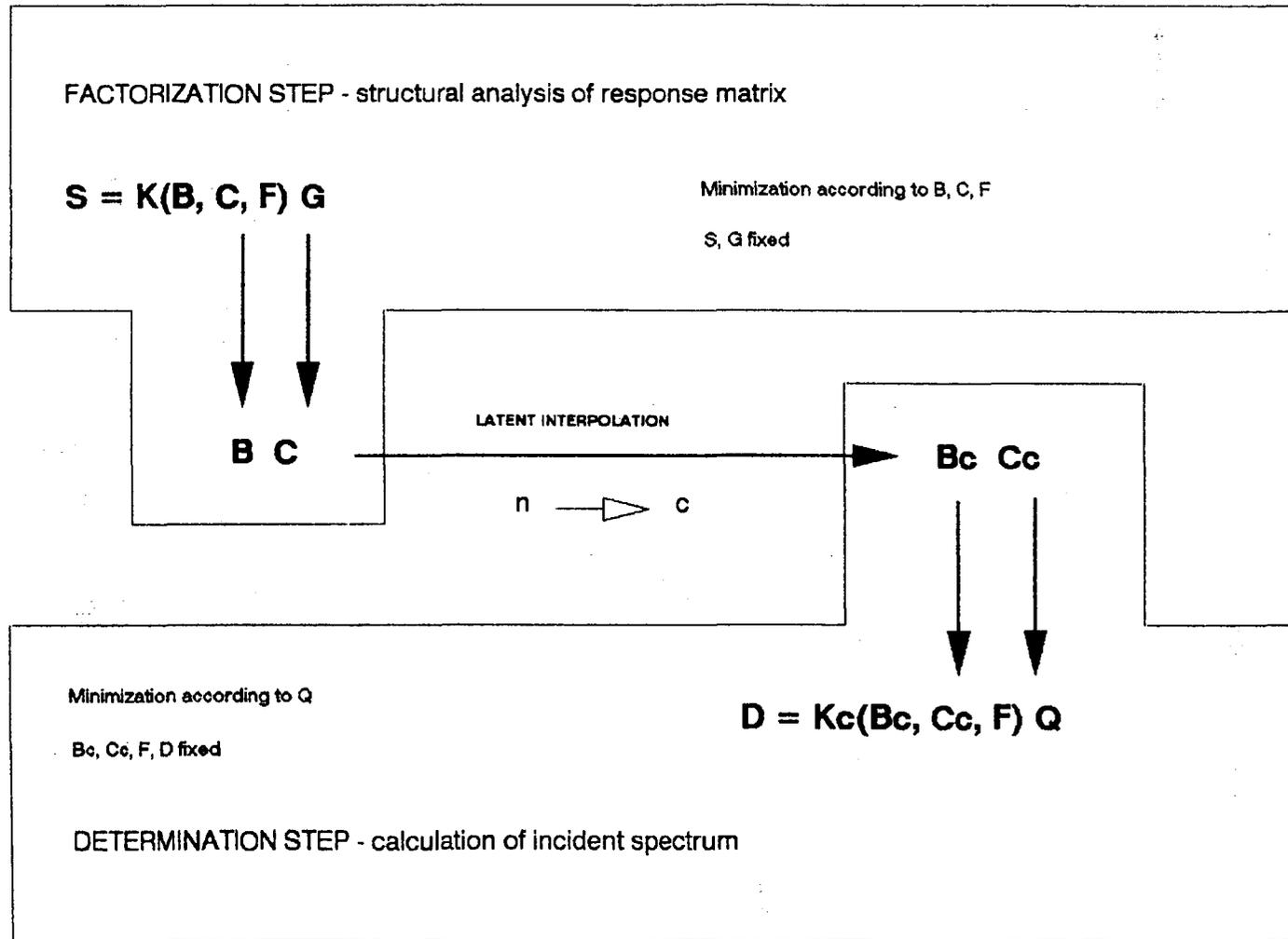
| Interference    | MSA [Bq]        |                            |                             |                            |                              |
|-----------------|-----------------|----------------------------|-----------------------------|----------------------------|------------------------------|
|                 | background only | 100 Bq of $^{60}\text{Co}$ | 200 Bq of $^{152}\text{Eu}$ | 13 kBq of $^{60}\text{Co}$ | 219 kBq of $^{152}\text{Eu}$ |
| Integral method | 6,4             | 535                        | 1 286                       | 69 735                     | $>10^6$                      |
| PNA method      | 2,6             | 5,3                        | 5,4                         | 53                         | 136                          |
| SCFA method     | 0,7             | 0,85                       | 0,89                        | 5,4                        | 18                           |
| PNA/SCFA ratio  | 3,71            | 6,24                       | 6,07                        | 9,9                        | 7,6                          |

The SCFA method is very advantageous to use, for instance, in ultra low-level g-spectrometry where counting rates in full energy peaks are extremely low as compared with background interferences. It enables to increase of the sensitivity by 5-10 times in comparison with the traditional full energy peak net area method.

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Figure 1. Factorization and determination in semiconductor spectrometry



## EVALUATION OF GAMMA RAY FIELDS BY HPGE SPECTROMETRY IN SITU

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### Introduction

A new method for calculation and evaluation of the semiconductor gamma-ray spectra is now proposed [1, 2]. The method is based on factorization of the response operator by a confirmatory least squares technique of factor analysis (SCFA). The SCFA approach is very advantageous for a construction of the relevant response operator for many spectrometric applications. A whole-spectrum processing in the response operator application allow us to expect mainly two advantages of this method: (a) increasing the sensitivity of the semiconductor gamma spectrometric method, and (b) the possibility of calculating the real incident (e.g. photon fluence rate) spectra. One can then comfortably accomplish an estimation of basic differential (spectral) and/or integral dosimetric characteristics.

### Instruments

Measurements have been performed using the conventional coaxial HPGe detector with a 12.5 % relative efficiency (in comparison to 3"x3" NaI full-energy PNA at 1.33 MeV) and a resolution of 1.9 keV (FWHM at 1.33 MeV). The non-perishable nature of this detector widens the application of Ge spectrometers to include field use of portable spectrometers. The useful energy of the detector is above 40 keV to more than 10 MeV. An experimental quantification of spectra by 256 channels and channel width  $\Delta E = 8$  keV has been utilized. It approximately corresponds to the gamma-spectra at 1.8 MeV.

The detector was cooling by the LN<sub>2</sub>. However, the *in situ* spectrometric measurement for application in gamma radiation dosimetry requires portability and flexibility in use. In order to allow operation of the detector in any orientation without LN<sub>2</sub> spillage, a multi-attitude cryostat (MAC) has been used which consists of a Dewar with LN<sub>2</sub> capacity of 7.0 litres and a holding time of 5 days. This allows the Dewar to be operated in the horizontal position, pointing vertically upward or vertically downward, without loss of LN<sub>2</sub>.

The MAC detector has been positioned in a 4 $\pi$  goniometer and, therefore, is movable to any measurable angle. Pulses from the detector have been fed into a portable multichannel analyzer (Canberra 35+) with connection to a PC/AT compatible computer system.

### Methods

If assuming that the physical spectra are given and the complete form of the response operator is known (see SCFA procedures in Ref. [1]), then an application has been described of the complete response operator  $K(c \times c)$  to measured discrete and/or continuous physical spectra put in the  $c \times r$  experimental matrix  $D$  as columns as follows

$$D = K Q, \quad (1)$$

where the  $c \times r$  matrix  $Q$  consists of the  $r$  unknown incident spectra as columns.

The photon fluence rate  $\phi$  [s<sup>-1</sup>.cm<sup>-2</sup>] from a point isotropic source characterized by an intensity  $Q$  [s<sup>-1</sup>] may be expressed in a distance from the source, given by  $R$  [cm], as follows

$$\phi = \frac{Q}{4\pi R^2}, \quad (2)$$

or as spectral photon fluence rate

$$\phi(E) = \frac{Q(E)}{4\pi R^2}, \quad (2a)$$

where  $Q(E)$  characterizes a spectral distribution of source intensity according to photon energy. In regard to the matrix approximation of spectra used in the spectrum measure description 1, we have analogously introduced a matrix equation for *in situ* measurement using definition 2a as follows

$$D = K Q = 4\pi R^2 K \Phi = K_F \Phi, \quad (3)$$

where  $K_F (c \times c)$  is the fluence rate response matrix and  $\Phi$  is the  $c \times r$  matrix with  $c$  vectors of the spectral fluence rates as columns corresponding to experimental quantification of the function  $\phi(E)$  by  $c$  points and  $R$  is the distance of calibration point from the detector.

Therefore, it has been found, in the aspect of SCFA performance, that solution of 1 could be very advantageously employed in the HPGe measurement *in situ*. As a matter of fact, certain difficulties are met in direct application of the SCFA solution in the *in situ* measurement which result from:

- (a) an inadequacy of the response operators such as  $4\pi R^2 K$  and  $K_F$ ,
- (b) a real (non zero) dimension of the coaxial HPGe detectors,
- (c) a Dewar flask shade of the detector.

Because the backscattering factor characterizes Compton scattering outside of the detector, it is becoming related to a concrete detection geometry of measuring and, therefore, it depends on a distribution of the photon fluence rate sources and on an arrangement of the surrounding material. Thus, if we omit this exterior factor in the reproduction of the response operator, a new reduced response operator  $\tilde{K}$ , or  $\tilde{K}_F$ , which describes only photon interactions in the detection medium, is that correct operator for the *in situ* application.

### Results and Discussion

In many of the *in situ* applications, the potential sources are mostly localized into sufficiently small spatial angles. If the angular localization  $\Delta\theta < 130^\circ$ , it is sufficient to perform only two measurements: for  $\varphi = 0^\circ$  and for  $\varphi = 180^\circ$  (the angle of the detector orientation). In this case, at least one of two measures is not shielded by the Dewar and gives correct values of the fluence. On the other hand, if there is no prediction on angular distribution of sources, some correction regarding the Dewar shade must be made [1].

Influence of the Dewar flask shading effect upon the *in situ* measurements may be seen by a comparison of the measured physical spectra in Figure 2, where are plotted the *in situ* physical spectra of some modelling source (see Figure 1) with "up" and "down" shading orientations of the detector. Corresponding incident photon fluence rate spectra for the vertically "upward" and "downward" orientations are shown in Figure 3.

Further, the main results and findings of present contribution may be summarized as follows:

1. A technique called the scaling confirmatory factor analysis (SCFA) presented elsewhere [2] can be advantageously employed for determination of the response operator characterizing an influence of measuring device on physical gamma-spectra obtained. The *in situ* response operator has been reproduced only from the internal factors of appropriate latent structure that do not depend upon materials surrounding the detector.

2. The photon fluence rate response operator for *in situ* application has been obtained from the reduced response operator by a correction according to the geometric factor  $4\pi(r_0+r)^2$ . The effective distance  $r_0$  has been determined via a performance of the radial calibration which yields a condition of, minimally, 10 cm distance of the detector cover from the potential sources.

3. The real incident gamma ray spectra achieved by application of the SCFA response operator allow direct evaluation of spectral distributions of the fundamental photon dosimetric quantities. For 5000 s counting time, the precision of present HPGe dosemeter did not exceed 0.1% of photon exposure and/or dose rate, and the  $^{137}\text{Cs}$  detection limit was about  $0.125 \text{ nGy}\cdot\text{h}^{-1}$  for 99% confidence level.

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Fig. 1. Spatial arrangement of modelling gamma sources

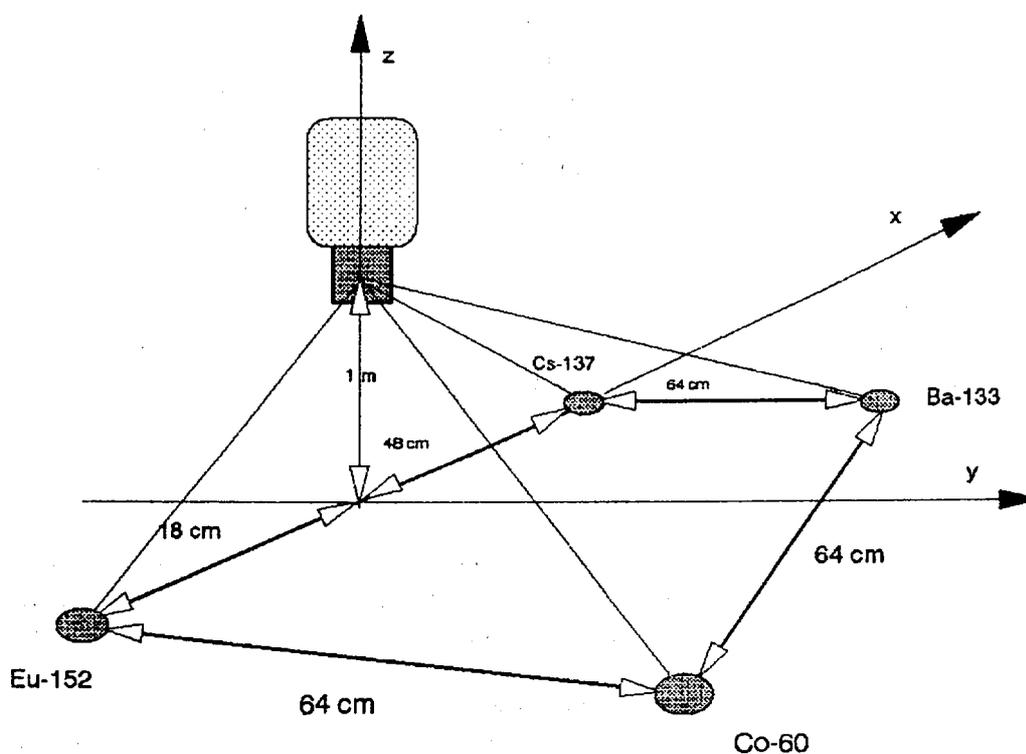


Fig.2. Experimental gamma ray spectra  
(1) without, (2) with shading, (3) backgr.

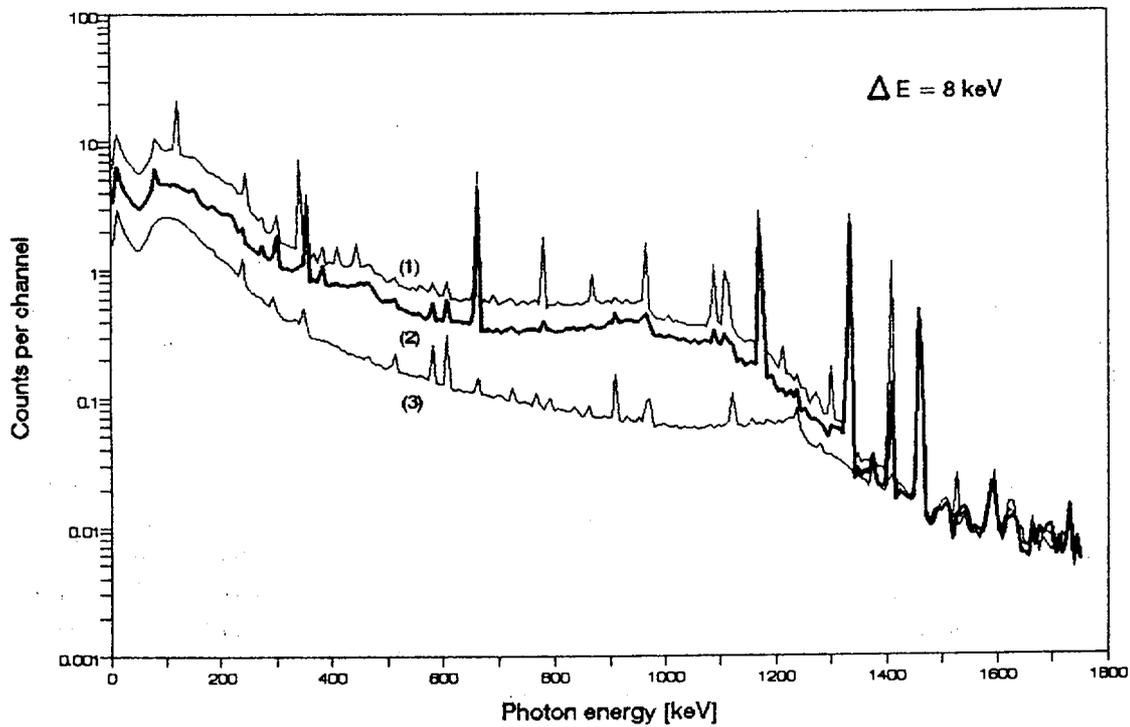
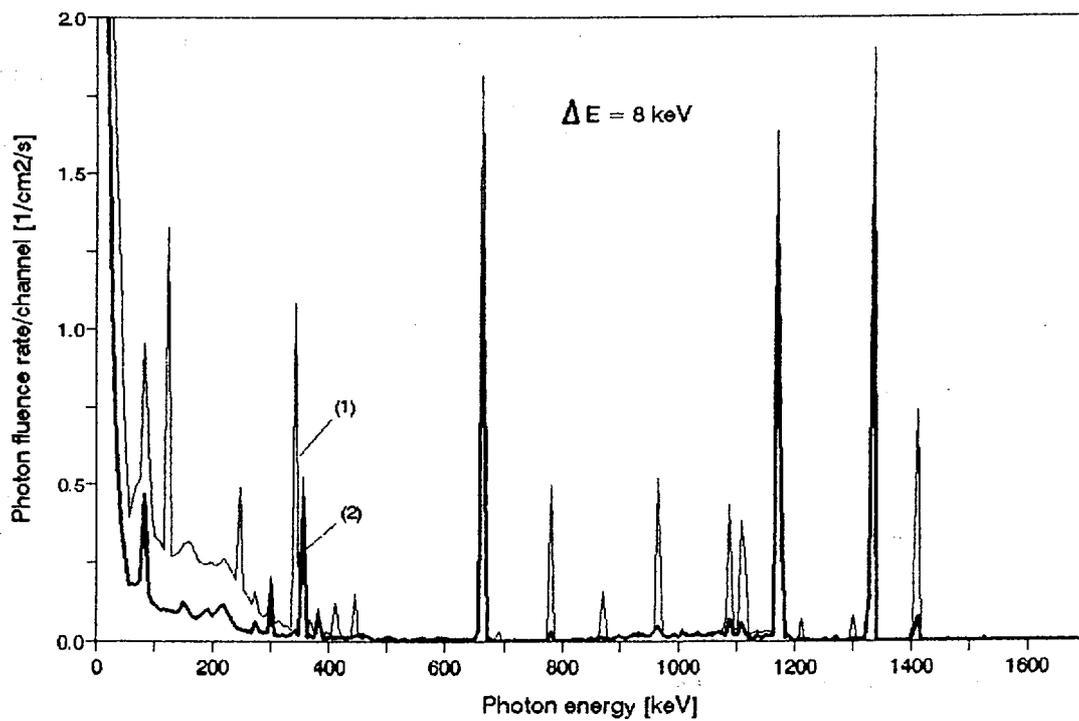


Fig.3. Incident gamma ray spectra  
(1) without, (2) with Dewar flask shading





## IN-SITU SPECTROMETRY OF $^{137}\text{Cs}$ IN THE SOIL BY UNFOLDING METHOD

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### Introduction

The in-situ gamma ray spectrometry is the method for rapid determination of a radionuclide concentration on and in the soil. Standard methods of in-situ spectrometry [1] are based on an assumption of gamma radiation attenuation by the soil. The standard methods require that assumption about the radiation attenuation by the soil have to be made in advance.

This contribution is aimed to the possibility of improving the in-situ gamma spectrometry to be independent on a knowledge about a depth distribution of  $^{137}\text{Cs}$  in soil and sufficiently sensitive for the measurement of the post-Chernobyl  $^{137}\text{Cs}$  at present, as well. The depth distribution of  $^{137}\text{Cs}$  averaged over a large area of soil is obtained by unfolding of the detector responses to primary and in soil forward scattered photons. The proposed method employs detector with and without collimator. The  $^{137}\text{Cs}$  distributions obtained from in-situ measurements are analysed, and comparisons are made to the results obtained with soil sampling and with standard in-situ spectrometry, as well.

### Methods

The detector response  $N(i)$  to  $i$ th characteristic of gamma ray field above ground with radionuclide distributed homogeneously in horizontal direction is described by integral equation :

$$N(i) = \int \sigma(i, \zeta) A(\zeta) d\zeta \quad i=1,2,\dots,n \quad (1)$$

where  $\sigma(i, \zeta)$  [ $\text{Bq}^{-1} \cdot \text{m}^2$ ] - detector response in gamma field of plane source of unit activity - per-area located at the depth  $\zeta$ ,  $A(\zeta) d\zeta$  [ $\text{Bq} \cdot \text{m}^{-2}$ ] - activity of the plane source to be determined,  $\zeta$  [ $\text{kg} \cdot \text{m}^{-2}$ ] - depth in the soil in units mass-per-area.

The unknown activity  $A(\zeta)$  in soil profile is obtained by unfolding of the Equation (1) [6].

The essential consideration of unfolding method is the linear independence of set of detector responses calculated  $\sigma(i, \zeta)$  (response matrix). The most promising and versatile unfolding code seems to be SAND II [7]. In practice, the good representation of activity distribution requires more steps in interval of soil depth that exceeds the number of the gamma field characteristics; as a consequence a first order approximation of the activity concentration in the soil profile must be introduced.

The response matrix is obtained by calculations in combination with experimental calibration.

In order to verify the reliability of the described method the comparison between laboratory measurements of soil samples and the analysis of the in situ recorded spectra is performed.

### Calculational procedure and results

The detector response  $\sigma(i, \zeta)$  to  $i$ th characteristic of gamma field with flux  $\phi(i, \zeta, \theta, E)$  per-unit-activity of plane radionuclide at the depth  $\zeta$  in the soil can be described as follows:

$$\sigma(i, \zeta) = \iint \phi(i, \zeta, \theta, E) \cdot R(i, \theta, E) d\theta \cdot dE \quad [\text{Bq}^{-1} \cdot \text{m}^2] \quad (2)$$

where  $R(i, \theta, E)$  - detector response in peak of total absorption for a parallel photon beam of energy  $E$  at angle  $\theta$ .

The Equation (2) can be simplified by assumption that the dependences  $R(i, \theta, E)$  are in the energy range of photons from 0.62 up to 0.662 MeV constant:

$$\sigma(i, \zeta) = \int \Phi(i, \zeta, \theta) \cdot R(i, \theta) d\theta \quad (3)$$

where  $\Phi(i, \zeta, \theta) d\theta$  - photon flux at angle  $\theta$  integrated over energy,  $R(i, \theta)$  - angular dependence of detector response for a parallel photon beam of energy 0.662 MeV.

The photon flux  $\Phi$  has been calculated by our Monte Carlo code SOILSC.

Homogeneous and isotropic plane source of thickness 0.5 cm was simulated up to 85 g.cm<sup>-2</sup> of depths in the soil, starting with the plane source on the ground. The plane source radii were chosen 100 m to represent an infinitive plane soil contamination with radionuclide <sup>137</sup>Cs.

Photon interaction processes of photoelectric absorption and Compton scattering (8) had been taken into account in the Monte Carlo program.

#### Experimental calibration and response matrix

A portable N-type high purity germanium HPGe detector with relative efficiency of 12,5% and resolution of 1,7 keV for 1,33 MeV gamma rays has been used. The detector was supported by a tripod at a height 100 cm above the ground. The orientation of the detector is facing downward.

A cylinder-shaped has been used to modify the distribution of track lengths in soil of photons impinging to the detector surface. The collimator is made from of lead cylinder with the outer/inner diameter 19/10 cm and height 10 cm laying on aluminium cylindere with the outer/inner diameter 30/10 cm and height 2cm. The collimator was located on the detector axis at height of its bottom 100 cm above the ground.

The experimental callibrations were aimed to determinate the responses  $R(i, \theta)$  (in Equation 3) of collimated and uncollimated detector in the energy region 0.62-0.655 MeV and in peak of total absorption of the 0.662 MeV photons, as well, and to asses the background in the energy region 0.62-0.655 MeV due to photons from the natural sources.

The detector responses (in peaks of total energy absorption) to scattered and unscattered photons of the plane source <sup>137</sup>Cs in dependence of depth in mass-per-area calculated from Equation 3 are shown in Fig.1.

The detector responses to primary photons of energy 0.662 MeV are marked by a, and to scattered photons with energies 0.62 - 0.655 MeV are marked by b.

Index  $i=1$  is the case of the uncollimated detector,  $i=2$  is the collimated detector, front of the collimator was at the same distance 100 cm above the soil as the detector front.

The statistical accuracy of calculated detector responses depends on the source depth. For all results of the detector response to primary photons, the standard deviations were within 5%. The standard deviation of the detector responses to scattered photons with energies 0.62-0.655 MeV is up to 20% for plane source <sup>137</sup>Cs on the ground, 15% for source in the depths up to 3.5 g.cm<sup>-2</sup>, 10% for source in deeper depths and 20% at the depth under 30 g.cm<sup>-2</sup>.

The detector responses to various characteristics of photon field shown that they are clearly different from each other up to soil depth of about  $30 \text{ g.cm}^{-2}$  and therefore can be used as elements of the response matrix in the Equation (1).

#### **Application to post-Chernobyl $^{137}\text{Cs}$**

The photon spectra were recorded over lawns in Southern Slovakia. In-situ gamma spectrometry was performed with and without collimator at the height 100 cm above the centre of the square area used for soil sampling. The finiteness of lawn was taken into account by geometric factor calculated by SOILSC.

Figure 2 shows the pulse height distribution measured by the unshielded detector at a location on the lawn. In the lower part of the figure the detail of the spectrum in energy interval 0.5 - 1.0 MeV is shown. Exponential approximation of the spectrum in the interval 0.67 - 0.9 MeV, bold line in the figure, is used for assessment of background of measurement of 0.62-0.655 MeV photons scattered in the soil.

The activities of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  are sufficient to appear as discrete peaks in the measured in-situ spectra and the forward scatterings of 0.662 MeV photons in the soil can be evaluated.

Fig.3-5 show depth distributions of the  $^{137}\text{Cs}$  activity in soil measured in surrounding of Bratislava, the places were in distance 20 km from each other. The results obtained by presented method are compared to results by soil sampling. The relative standard deviation of the  $^{137}\text{Cs}$  activity in soil samples were  $<10\%$  for each depth of soil.

The downward transport of  $^{137}\text{Cs}$  have occurred to a peak concentration that lies below the surface of lawns. The differences between presented method and soil sampling of the distributions in Fig.3 and 4 are in 10% of relative standard deviation, except distributions at the deeper depths where resolution of unfolding method is worse. Unfolded distributions in Fig.5 is determined with large uncertainties because of relatively low activity of  $^{137}\text{Cs}$  in soil.

The  $^{137}\text{Cs}$  activity-per-mass and/or activity-per-area in soil determined by presented method of deconvolution, by sampling method and by standard in-situ spectrometry using the exponential depth distribution in the soil, that has been determined by exponential least squares fit of results of soil sampling, are given in Table 1.

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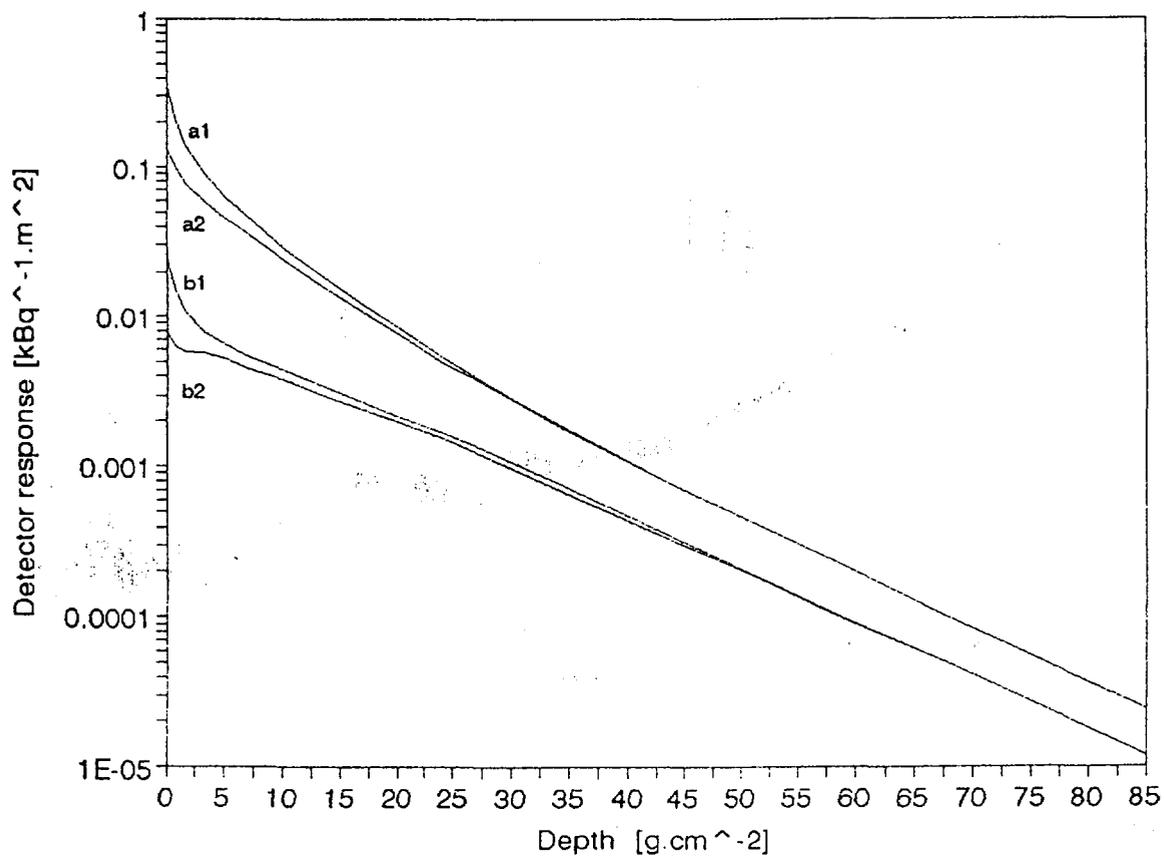


Fig.1 Response matrix of the in-situ equipment used in this paper. The lines  $a_1$ , and  $b_1$  are responses of the uncollimated detector to 0.662 MeV and to 0.62-0.655 MeV photons, respectively. The lines  $a_2$  and  $b_2$  are the same as  $a_1$  and  $b_1$ , but for the collimated detector.

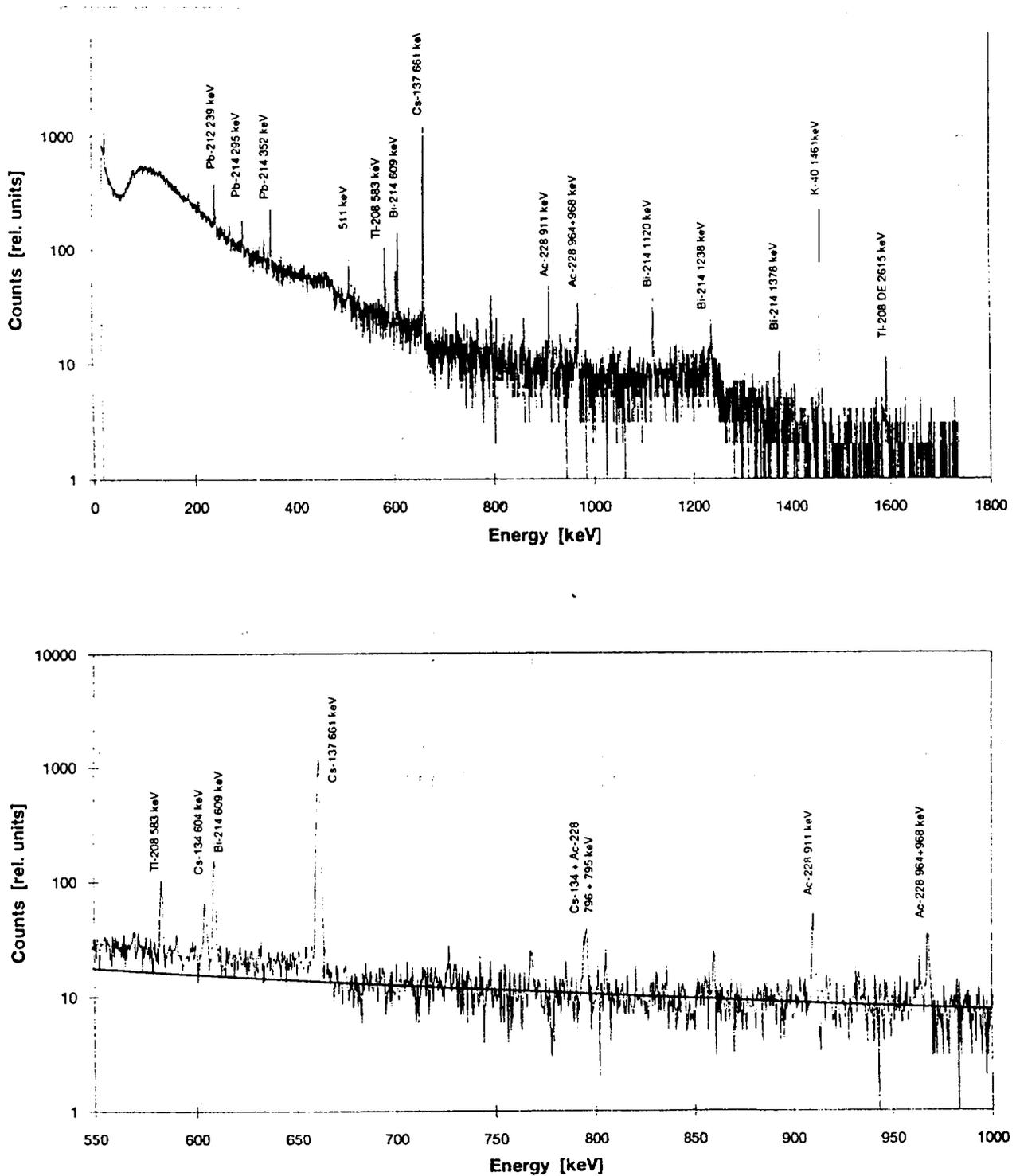


Fig.2 Impulse height spectrum as detected over a lawn in Southern Slovakia. At the bottom of the figure is the middle part of the spectrum in more detail. The bold line is an exponential approximation of the spectrum in the energy region 0.67-0.9 MeV.

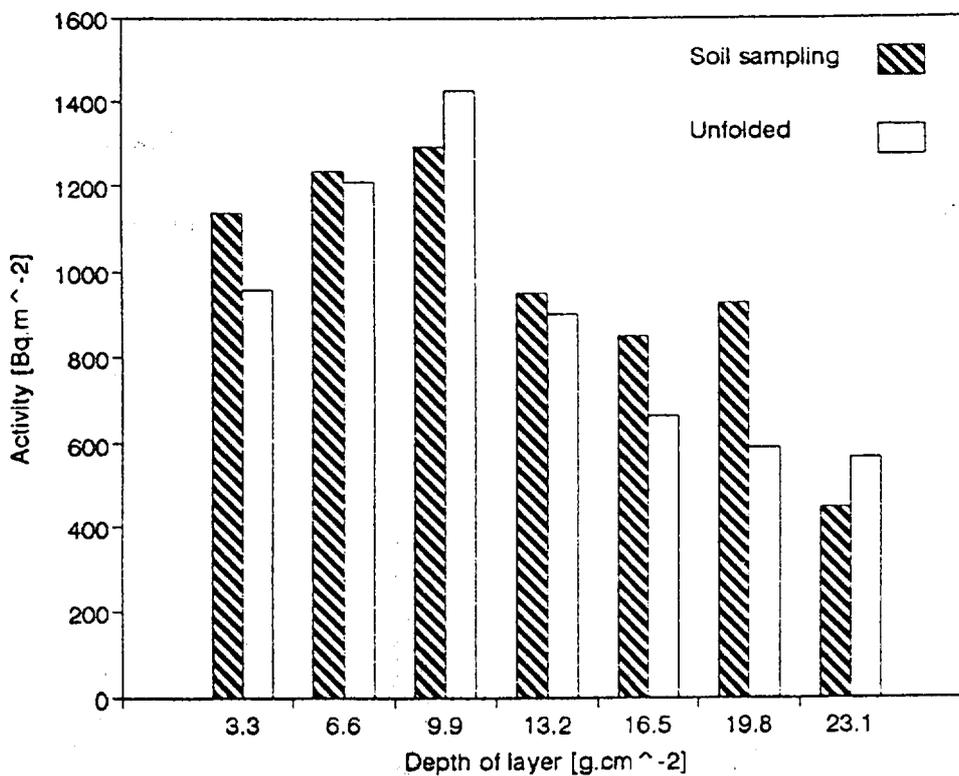


Fig.3 Distribution of the <sup>137</sup>Cs activity in soil profile of the lawn in the locality No.1 determined by presented method and by soil sampling, as well. The thicknesses of the soil layers were 3.3 g.cm<sup>-2</sup>. The soil consists of light black earth.

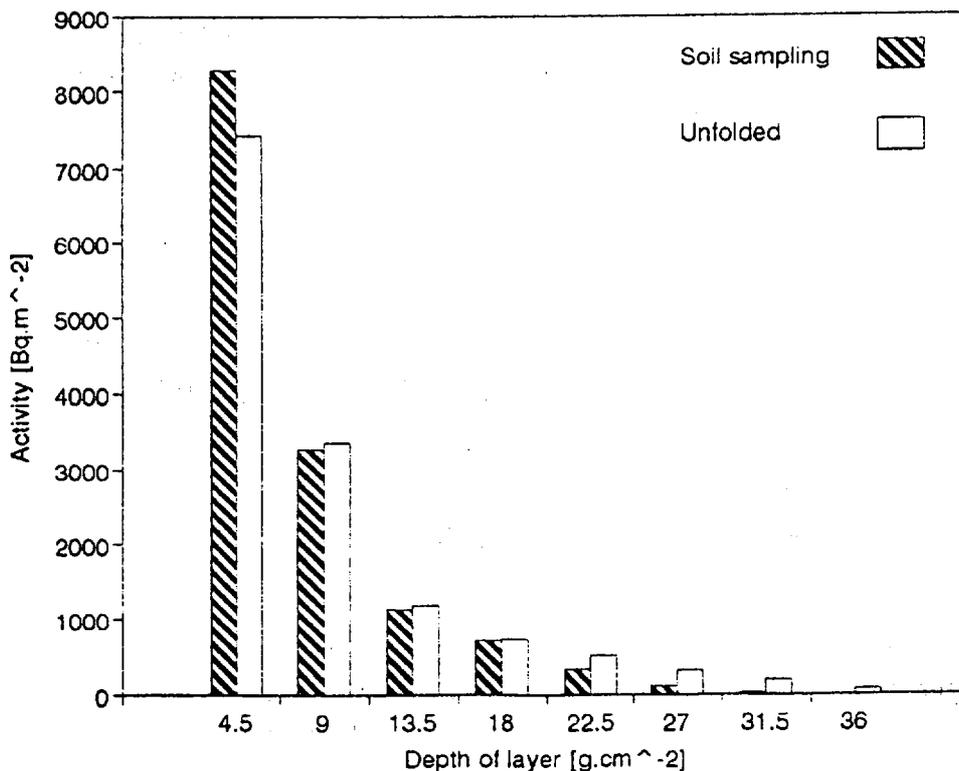


Fig.4 Distribution of the <sup>137</sup>Cs activity in soil profile of lawn in the locality No.2 determined by presented method and by soil sampling, as well. The thicknesses of the soil layers were 4.5 g.cm<sup>-2</sup>. The soil consists of clay.

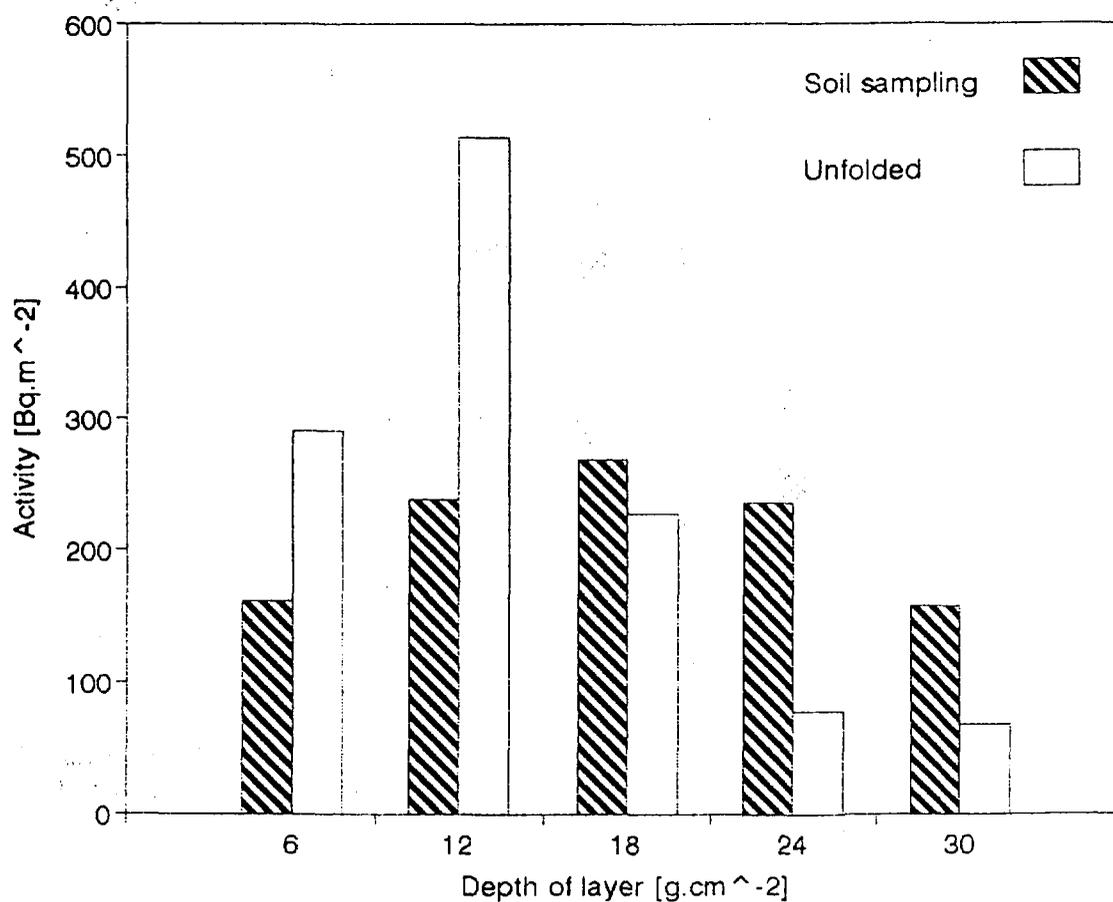


Fig.5 Distribution of the <sup>137</sup>Cs activity in soil profile of lawn in the locality No.3 determined by presented method and by soil sampling, as well. The thicknesses of the soil layers were 6.0 g.cm<sup>-2</sup>. The soil was mixture of clay and small stones.

| No. of locality | Presented method       |                        | Soil sampling          |                        | Standard method        |                        |
|-----------------|------------------------|------------------------|------------------------|------------------------|------------------------|------------------------|
|                 | [kBq.m <sup>-2</sup> ] | [Bq.kg <sup>-1</sup> ] | [kBq.m <sup>-2</sup> ] | [Bq.kg <sup>-1</sup> ] | [kBq.m <sup>-2</sup> ] | [Bq.kg <sup>-1</sup> ] |
| 1*              | 6.3±1.6                | 274±68                 | 6.8±0.7                | 258±26                 | -                      | 307                    |
| 2**             | 13.2±3.5               | -                      | 13.8±1.5               | -                      | 12.5                   | -                      |
| 3***            | 1.2±0.5                | 39±16                  | 1.1±0.2                | 36±7                   | -                      | 33                     |

Table 1. <sup>137</sup>Cs activity in soil of lawns in Southern Slovakia determined by the method presented in this paper, by the soil sampling and by the standard in-situ spectrometry using the depth distribution in the soil according to the results of the soil sampling.



## SPECIATION OF RADIOCESIUM IN ATMOSPHERIC AEROSOL AFTER THE CHERNOBYL ACCIDENT

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Physico-chemical forms of radiocesium released into the atmosphere during the nuclear power plant accident in Chernobyl can significantly influence its behaviour, above all resuspension and transport to other compartments of the environment, including pedosphere and hydrosphere.

Hilton et al. [1], analyzing  $^{137}\text{Cs}$  forms in atmospheric aerosol collected in May, 1986 at Oxfordshire location (U.K.), found out marked differences as compared with the results of the analysis of samples collected in 1959 which contained  $^{137}\text{Cs}$  from nuclear weapon tests. A difference in water-soluble  $^{137}\text{Cs}$  fraction was the most distinct, amounting to 72% as compared to only 8% in the stored samples.

Bobovnikova et al. [2], however, found out in the analysis of fallout material collected at a distance of 18 km from the NPP Chernobyl during the period of the accident 26 April - 3 May 1986 that for  $^{137}\text{Cs}$  insoluble form was mainly typical. The content of water-soluble radiocesium fraction was about 9% and in the studied period ranged from 2.6 to 19%. Jansta [3] found in the analysis of the fallout material collected in Košice (Slovak Republic) in the period 29 April - 21 May 1986 the soluble fraction to be less than 10% of the total  $^{137}\text{Cs}$  activity in the fallout.

On the basis of the mentioned different results we decided to analyze forms of radiocesium on aerosol filters exposed in Prague in the period after the Chernobyl accident [4]. The aim of this analysis was to verify the hypothesis that physico-chemical forms of radiocesium in the fallout after the accident could depend on the transport conditions, including the distance of a sampling location from Chernobyl.

### Experimental

Four aerosol filters (marked A-D) exposed in the period of direct contamination (A-30/4/86, B-1/5/86, C-4/5/86, D-6/5/86) were analyzed. The set was completed with filter E which was exposed only after the direct contamination (12/5/86 to 19/5/86) had finished. The used procedure of sequential analysis was based on the method proposed by Tessier [5] for the study of soils and modified for the analysis of aerosol filters according to Hilton et al. [1]. The  $^{137}\text{Cs}$  activity in the samples of individual steps of sequential analysis was determined by gamma spectrometry using GeLi detector. Experimental procedure and measurement technique are described in detail elsewhere [6].

### Results

The results of the sequential analysis of  $^{137}\text{Cs}$  forms in the samples of atmospheric aerosol collected in Prague in the period of direct contamination by Chernobyl plume are summarized in Table 1. For comparison also results are given of the sample E from a later period and the results adopted from [1] concerning atmospheric aerosol collected at Oxfordshire (U.K.) in May, 1986. The  $^{137}\text{Cs}$  activity of individual sequential analysis fractions is expressed as a percentage of the total  $^{137}\text{Cs}$  activity in the analysed sample.

From the results it is obvious that the prevailing form in all samples taken in the period of direct contamination was water-soluble radiocesium. The observed values, ranging from 40 to

44%, are, however, lower when compared with the results given by Hilton et al. who found 72% of soluble  $^{137}\text{Cs}$ .

**Tab. 1** Results of a sequential extraction analysis of radiocesium in atmospheric aerosol samples (percentage of the total activity).

| Step | Extractant  | A  | B  | C  | D  | E  | UK* |
|------|---|----|----|----|----|----|-----|
| 1    | H <sub>2</sub> O  | 43 | 44 | 49 | 40 | 30 | 72  |
| 2    | 1 M MgCl <sub>2</sub> (pH 7)  | 14 | 21 | 16 | 26 | 16 | 0.5 |
| 3    | 1 M NH <sub>4</sub> Cl  | 3  | 4  | 4  | 7  | 3  | 15  |
| 4    | 1 M NaOAc (HOAc, pH 5)  | N  | 2  | N  | 6  | 1  | 3   |
| 5    | 0.04 M NH <sub>2</sub> OH.HCl in 25% HOAc                                 | N  | 2  | N  | 8  | 2  | 2   |
| 6    | 0.1 M HCl   | 2  | N  | 3  | N  | N  | N   |
| 7    | 3 vol. 0.02 M HNO <sub>3</sub> + 5 vol. 30% H <sub>2</sub> O <sub>2</sub> | 12 | 2  | 13 | 11 | 6  | 2.5 |
| 8    | Residue   | 26 | 25 | 15 | 2  | 43 | 5   |

\* taken from the publication by Hilton et al.

N the solution was not applied

The second most important fraction was formed by undissolved residues in which the content of  $^{137}\text{Cs}$  ranged from 2 to 26% and gradually decreased with time from the accident. Another significant fraction was found after leaching with MgCl<sub>2</sub> solution, i.e. in the fraction subject to ion exchange. This fraction ranged from 14 to 25% and probably did not depend on the time of sample collection and also differs markedly from the results published by Hilton et al.

The results of analysis of sample E confirmed the expected increase in radiocesium content in the undissolved residue and the decrease of water-soluble fraction because after direct contamination had finished, resuspension became the main contamination source of ground-layer atmosphere. In resuspension, however, insoluble cesium forms are preferred because they stay on the surface and do not penetrate into deeper soil layers. In addition, the increase of insoluble  $^{137}\text{Cs}$  fraction in resuspended material is influenced by interaction of soluble radiocesium with soil complex which results in firm bond of radiocesium with soil particles and its immobilization in a surface soil layer [7].

From the presented results it can be concluded that physico-chemical forms of radiocesium in atmospheric aerosol and fallout after the NPP accident at Chernobyl as well as particulate size distribution can depend on the distance or the conditions of transport from a contamination source to a sampling location. The influence of the conditions of radiocesium transport could result in the observed differences in the  $^{137}\text{Cs}$  penetration into soil profile in different locations and also in the found dependence of the resuspension factor for  $^{137}\text{Cs}$  on the level of its fallout in the period of NPP accident for different locations in Europe [8].

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## INDOOR RADON EPIDEMIOLOGICAL STUDY

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### Introduction

In 1991, under the co-ordination by the former Centre of Radiation Hygiene of the National Institute of Public Health in Prague, an epidemiological study, designed by the late Josef Ševc, was started. The study subject was lung cancer incidence among inhabitants of a specific locality in the middle of a larger area of granitoids, carrying a geological term Middle-Bohemian Pluton.

The locality, mostly agricultural, exhibits considerable geological disturbances and the inhabitants are exposed to different, prevailingly high levels of radon in the dwellings. The houses situated on the lines of disturbances exhibit very high levels of radon-222 and its daughter products, while in the adjacent houses outside the lines, the concentrations of radon are often manifold lower, still being higher than the average for the country.

### Description and objectives

The study is a long-term prospective (non-parallel) cohort study of lung cancer and possibly other causes of death. The methodological approaches are identical to those applied in Czechoslovak occupational studies of uranium miners (Ševc et al, 1991). The study population include inhabitants of the area, who had resided there for at least three years and at least one of these between 1.1.1960 and 31.12.1989. A total of 11865 inhabitants satisfied these criteria.

Data on vital status of each cohort member, residences during different time periods, smoking habits, occupational and other exposures have been collected through interviews of residents or relatives at home during which also data on constructional changes of buildings were registered. Information on vital status and causes of death was collected and/or verified at the local authorities, in the national population registry, in health documentation of the corresponding district hospital or oncological facilities and their pathologico-anatomical departments (with regard to histological characteristics of the tumor).

The cumulative exposure of each respondent is being assessed on the basis of measurements in dwellings, time spent there and estimation of previous exposure levels by a model accounting for constructional changes in buildings. One year lasting measurements of radon daughter products by integral dosimeters (Kodak film LR 115) were performed in practically all dwellings of the specified area.

A model relating radon concentrations to radon ingress rate from the soil and ventilating conditions has been introduced and was with positive results tested in this and the Jáchymov area, permitting to estimate the influence of performed building changes on the consequent residential radon concentration. A study of possibilities in improvement of cumulative exposure assessment by determination of Pb-210 in glass objects in the dwellings and by measuring Pb-210 in persons with parallel determination of Pb-210 intakes is being started.

### Preliminary results

Table 1. summarizes radon measurements in houses in term of equivalent equilibrium concentration and compares them with the results of a pilot study in Petrovice in 1990-91 which gave the stimulus for the epidemiological study.

**Tab. 1** Distribution of radon concentrations

| EEC Rn (Bq/m <sup>3</sup> ) | Petrovice | Study area |
|-----------------------------|-----------|------------|
| 10 - 199                    | 22 %      | 45 %       |
| 200 - 499                   | 30 %      | 41 %       |
| 500 - 999                   | 32 %      | 12 %       |
| 1000 -                      | 16 %      | 2 %        |

The overall mean of 300.4 Bq/m<sup>3</sup> for the study area based on year measurements in practically all houses (N=2480) confirmed substantially higher concentrations than the country mean EEC 40-80 Bq/m<sup>3</sup>. However, the levels are not uniform, nearly half the houses are below the EEC level of 200 Bq/m<sup>3</sup> (Tab.1) and the maximum found concentration was 11 kBq/m<sup>3</sup>. By the end of 1991, a total of 2711 deaths have been registered in the cohort. In 2607 cases (96% of deaths) the causes of death have been identified. A total of 143 cases of lung cancer (5% of all deaths) have been registered by 1994.

**Tab. 2** Cause specific mortality by 1991

| Sex | All deaths |      | Lung cancer |      | Other cancers |      | Violent deaths |      | Other causes |      | Un-known |
|-----|------------|------|-------------|------|---------------|------|----------------|------|--------------|------|----------|
|     | O          | O/E  | O           | O/E  | O             | O/E  | O              | O/E  | O            | O/E  |          |
| M   | 1530       | 0.96 | 131         | 1.08 | 96            | 0.67 | 96             | 0.67 | 1035         | 0.96 | 48       |
| F   | 1181       | 0.89 | 12          | 0.78 | 36            | 0.53 | 36             | 0.53 | 869          | 0.88 | 56       |
| Tot | 2711       | 0.93 | 143         | 1.04 | 132           | 0.62 | 132            | 0.62 | 1904         | 0.92 | 104      |

Table 2 displays the distribution of death causes and the ratio of observed (O) to expected (E) cases among collected death cases in the cohort, generally, somewhat lower ratios than one reflect the non-industrial character of the region, with the exception of lung cancer in man.

The differences in the O/E ratios for lung cancer among the separate communities, given in Table 3, indicate that even in the situation of generally lower mortality, the dependence of lung cancer mortality on radon exposure cannot be excluded.

The study is being continued in completing the data and thorough analyses of the lung cancer risk in relation to individual cumulative exposure. A nested-in case control study, to which collection of additional data has already started, will be performed.

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Table 3: Radon characteristics (EEC) and mortality by communities

| Community  | Rn concentration<br>Bq/m <sup>3</sup> |       | Lung ca |      | All deaths |      |
|------------|---------------------------------------|-------|---------|------|------------|------|
|            | Mean                                  | Max   | 0       | 0/E  | 0          | 0/E  |
| Petrovice  | 463                                   | 11165 | 24      | 0.89 | 571        | 0.98 |
| Kovářov    | 330                                   | 3552  | 16      | 1.13 | 242        | 0.80 |
| Kostelec   | 322                                   | 2642  | 21      | 1.79 | 226        | 0.87 |
| Hrazany    | 351                                   | 1737  | 8       | 1.20 | 133        | 0.97 |
| Záhořany   | 261                                   | 1470  | 8       | 0.76 | 181        | 0.85 |
| Chyšky     | 225                                   | 1261  | 32      | 1.31 | 515        | 0.98 |
| Zhoř       | 205                                   | 1032  | 3       | 0.42 | 146        | 0.96 |
| Hrejkovice | 237                                   | 864   | 8       | 0.79 | 204        | 0.91 |
| Dmýštica   | 208                                   | 911   | 8       | 0.95 | 177        | 1.07 |
| Kučer      | 164                                   | 920   | 15      | 0.90 | 316        | 0.88 |



## RELATIVE RISK MODELS OF LUNG CANCER IN URANIUM MINERS

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### Introduction

Studies of underground miners of uranium and other substances are at present the principal source of information on the long term effects of exposure to radon and its progeny. One of the largest such studies is that of uranium miners in West Bohemia (Jáchymov). This study, sometimes referred to as the S cohort, was set up in 1970 by the late Josef Ševc.

### Methods

The study population of the S cohort involve uranium miners, that started underground work at the Jáchymov and Horní Slavkov mines in the period 1948-59, and had worked for at least four years. A total of 4320 men satisfied these criteria.

During the decade up to 1990, follow-up of the cohort mainly relied on the national population registry. In order to improve the follow-up, a series of additional checks were conducted: in the files of the Czech and Slovak Pensions Offices, by local enquiries, and by direct correspondence (Tomášek et al, 1994b). These additional efforts resulted in an increase of more than 10% in the numbers of men known to have died or emigrated.

An exceptional feature of the S study is the large number of measurements of radon concentrations made in each mine-shaft (mean number per year and shaft was 223 in the period 1949-60). Each man's annual exposures to radon progeny in terms of working levels were estimated combining measurement data with the men's employment details. Recent data revisions revealed that for some of the men (about 10%), exposures at other Czech mines had not previously been taken into account (Tomášek et al, 1994a). In the most recent revision of exposures, appropriate adjustments were made for individual job categories (miners, other manual professions, supervisors, and exploratory workers).

The excess relative risk models were used in the form

$$RR = c ( 1 + ERR(w,x) ),$$

where ERR is excess relative risk, w and x denote exposure history and modifying variable, and c is an intercept term that allows the mortality rate for "unexposed" cohort to differ from that in the general population.

### Results and discussion

The increased mortality (O/E=1.58) in the cohort is largely affected by mortality from lung cancer. Nevertheless the mortality from violence and accidents is also increased, namely in the first part of follow-up, whereas the mortality from other causes increases after 20 years since first exposure (Tab.1).

A similar time pattern is observed in some groups of diseases generally associated with tobacco consumption (Tab.2). When the cohort was identified in 1970, information on the smoking habits in the cohorts could not be recorded. As the increase in mortality from cancers other than lung cancer is not likely to be associated with radon exposure (Darby et al, 1995), the data indicate that the smoking in the S cohort might be different from the general population.

**Tab.1** Time and cause specific mortality

| Time since 1st exposure | Lung cancer | Violent deaths | Other causes |
|-------------------------|-------------|----------------|--------------|
| 4- 4.9                  | 2.56        | 3.54 *         | 0.26         |
| 5- 9.9                  | 5.54 *      | 1.41 *         | 0.86         |
| 10-14.9                 | 9.28 *      | 1.58 *         | 1.10         |
| 15-19.9                 | 8.34 *      | 1.87 *         | 1.09         |
| 20-24.9                 | 6.06 *      | 1.18           | 1.34 *       |
| 25-29.9                 | 4.49 *      | 1.39           | 1.17 *       |
| 30-34.9                 | 3.36 *      | 1.39           | 1.17 *       |
| 35-                     | 2.72 *      | 1.13           | 1.23 *       |

The asterisk denotes one-sided test at 5% significance.

**Tab.2** Time and cause specific mortality

| Cause                | Time since 1st exposure |       |       |       |
|----------------------|-------------------------|-------|-------|-------|
|                      | 0-9                     | 10-19 | 20-29 | 30-   |
| Respiratory diseases | 0.35                    | 1.07  | 1.42* | 1.27* |
| Circulatory diseases | 0.86                    | 1.10  | 1.24* | 1.12* |
| Cancers excl lung    | 0.81                    | 0.93  | 1.15  | 1.28* |

Most of the miners' studies demonstrated the linear relationship between relative risk and cumulative exposure (Lubin et al, 1994). In a simple model in which the excess relative risk increases linearly with total 5 year lagged cumulative exposure, two parameters are estimated: the coefficient of ERR/WLM and intercept. The overall ERR/WLM estimate 0.013 for the most recent data is higher than the comparable estimates previously reported for the study: 0.0034 - (Lubin et al, 1994) and 0.0064 (Tomášek et al, 1994b). The corresponding intercept is 2.43 (CI:1.73-3.41).

Factors known to influence the excess relative risk per unit exposure were analyzed. The strong influence of time since exposure (TSE) was found. In addition, the effect of exposure rate was investigated, originally, in three categories (0-1.9, 2-3.9, 4+), but as there were no differences between the first two categories in the analyses, these were combined. Therefore, cumulative lagged exposures in the three TSE windows were split into two windows according to their concentrations (0-3.9, 4+). It was found that the ERR/WLM estimate corresponding to higher exposure rates was significantly lower. The effect of age, that was found in almost all miner studies (Lubin, 1994), was investigated in the same way as the other exposure modifiers, i.e. by the methods of windows. The estimates for the model with all the exposure modifiers are given in Tab.3. The confidence interval for the intercept in the last model is narrower in comparison to other models.

Further investigations of the intercept showed that when age factors were present in the model as continuous variables, the age-dependent intercept remained nearly constant (close to 1.5) suggesting that in the absence of exposure to radon, the estimated mortality from lung cancer in the cohort would be about 1.5 times higher than in the general population. If this estimated baseline lung cancer mortality in the study were true, the ERR/WLM coefficient related to simple cumulative exposure would be about 0.015 (95%CI: 0.0135-0.0165).

**Tab.3** Effect of TSE, exposure rate, and age

|            | Estimate | 95% CI      | Chi-Sq | DF |
|------------|----------|-------------|--------|----|
| Intercept  | 1.46     | 1.00 2.14   |        |    |
| ERR/WLM    | 0.124    | 0.077 0.199 |        |    |
| TSE:       |          |             | 69.53  | 2  |
| 5-14       | 1        |             |        |    |
| 15-24      | 0.26     | 0.11 0.41   |        |    |
| 25-34      | 0.07     | 0.02 0.13   |        |    |
| Exp.rate:  |          |             | 8.69   | 1  |
| 0-3.9      | 1        |             |        |    |
| 4-         | 0.47     | 0.21 0.72   |        |    |
| Age-at-exp |          |             | 52.07  | 2  |
| -29        | 1        |             |        |    |
| 30-39      | 0.60     | 0.36 0.85   |        |    |
| 40-        | 0.28     | 0.18 0.38   |        |    |

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## OUTLINES OF A STUDY OF SOME INDICATORS OF EXPOSURE OF UNDERGROUND WORKERS TO RADON IN SLOVAKIA

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Epidemiological studies performed on people professionally exposed in uranium and non-uranium hard-rock mines showed enough consistent data about risk of lung carcinomas caused by irradiation of lung tissue by radon and its daughter products. A thorough study was performed in the last two decades in the Czech republic (Sevc et al. 1976, 1984, 1988, 1993). Measurements of the radon in non-uranium mines and caves in Slovakia showed that radon might be a considerable occupational risk factor for underground workers there employed (UNSCEAR 1988).

There is no consistent information about the health consequences and about the magnitude of risk of lung cancer in Slovak underground workers, therefore a study was designed to give bases for such information.

Chromosomal aberrations (CA) serve contemporarily as good detectors of impairment of the human organism by ionizing radiation. The numbers of CA in peripheral blood lymphocytes depend from the absorbed radiation dose. After an acute whole-body irradiation it is possible to estimate the absorbed dose if this is not known (IAEA 1986). There are scientifically based hypotheses about the causal association of chromosomal aberrations and radiation cancer (UNSCEAR 1988) so that the CA may serve as indicators of genotoxicity.

The projected study has a dose-effect scaffold and its projected goals are on the side of dose assessment:

- 1) to determine the professional exposure of underground workers to radon by personal dosimetry and to establish a databasis of integrated personal doses for future epidemiological studies.
- 2) to introduce the method of counting the radionuclide  $^{210}\text{Pb}$  in the bones and skulls of underground workers.

On the side of effect assessment the goals are :

- 1) to find out if a dependence of the number of CA and micronuclei in the lymphocytes from radon dose exists. The measurements of radon in mines, preferentially those by personal dosimeters as well as an existing database of radon concentration measurements in dwellings and caves will serve for the radon dose estimate.
- 2) to compare the DNA-repair rate in the lymphocytes of people professionally exposed to radon in Slovak caves with the same rate as assessed in unexposed people.
- 3) to find out the proportional role of confounding factors such as smoking, alcohol abuse, exposure to dust etc., which might influence upon the interpretation of the dose from radon to health response relationship.
- 4) to establish a central database of lung cancer cases in professionally exposed underground workers for future epidemiological studies.

The experimental design to assess the dose will be following :

The magnitude of the exposure of underground workers to radon and its daughters will be estimated either indirectly or directly.

The indirect method relies upon measuring the concentration of radon using methods introduced by N.I.O.S.H. (1987). Continual and discontinual measurements of radon and its daughters in the underground workplaces will be performed. As the most reliable method of assessing the personal dose, the personal dosimetry will be introduced.

The direct method will rely upon the determination of miners' radiation burden based on the  $^{210}\text{Pb}$  activity deposited in skeleton (Laurer et al. 1993). The radionuclide  $^{210}\text{Pb}$  is retained in human skeleton with an effective half-life of 15 years. By using a suitable model it is possible to estimate the exposure of subjects during the whole working period from measured  $^{210}\text{Pb}$  activity.

The experimental design to assess the effects will have three main methodological parts :

- i) determination of the count of chromosomal aberrations and micronuclei using classical methods;
- ii) determination of the chromosomal exchanges using the fluorescent in situ hybridization method;
- iii) determination of the DNA-repair rate by UDS.

Donors of blood will be invited upon an agreed consent from the population of inhabitants whose dwellings have been measured for radon concentration. A questionnaire will be filled with every donor, it should provide data which will serve for the estimation of the radiation body burden from radon. There will be also a search for data necessary for getting information about confounding factors which may influence the CA counts (smoking, exposure to other clastogens etc.)

The part i) will consist of following methods :

A sample of blood will be stimulated by a mitogen, cultivated in vitro, and preparations for cytogenetic evaluation will be made. The numbers of chromosomal aberrations and micronuclei will serve as the endpoints followed.

The preparations for counting the chromosomal aberrations will be made 48 hours and the preparations for counting the micronuclei 72 hours from the begin of cultivation in vitro in binucleated cells after a telophase arrest performed with cytochalasin B.

The part ii) will consist of a method which should be introduced as new to this laboratory and as such it must be learned.

The part iii) will consist of following methods :

Lymphocytes from a sample of blood will be separated and irradiated by ultraviolet light by a standard procedure. Thereafter the cells will be incubated in vitro with tritium labelled DNA precursors. The rate of incorporation will be measured by scintillation counting and it will serve as an indicator of the DNA-repair rate in the sample.

The results of counting will be divided into groups according to the quartiles of the radiation body-burden distribution and all confounding factors (i.e. smoking). The biometric evaluation will consist of tests of differences between the counts of CA belonging to various groups of probands. Also, the differences among group-sums of CA will be tested. The dependence of CA-counts from the radiation body burden will be tested by regression procedures.

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## ATTEMPT TO DETERMINE RADON ENTRY RATE AND AIR EXCHANGE RATE VARIABLE IN TIME FROM THE TIME COURSE OF INDOOR RADON CONCENTRATION

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### Introduction

For radon diagnosis in houses the "ventilation experiment" is used as a standard method. After removal of indoor radon by draught the build-up of radon concentration  $a(t)$  [Bq/m<sup>3</sup>] is measured continuously and from the time course the constant radon entry rate  $A$  [Bq/h] and the exchange rate  $k$  [h<sup>-1</sup>] is calculated by regression analysis using the model relation  $a(t) = A(1-e^{-kt})/kV$  with  $V$  [m<sup>3</sup>] for the volume of the room. The conditions have to be stable for several hours so that the assumption of constant  $A$  and  $k$  are justified. During the day both quantities are independently (?) changing, therefore a method to determine variable entry rate  $A(t)$  and exchange rate  $k(t)$  is needed for a better understanding of the variability of the indoor radon concentration.

### Approaches to the time analysis of the radon concentration

The variation in time of the radon concentration is studied by the linear (compartmental) model

$$a(t)' = -k(t) a(t) + A(t)/V,$$

expressing the balance of removal and entry rate by the derivation  $a^*$  [Bq/(h m<sup>3</sup>)]. This differential equation has a general solution which can be used in principal for the determination of the unknown functions  $A(t)$  and  $k(t)$  by numerical methods but it seems to be very complicated and unrealistic. The other way is to use the differential equation itself.

### Deterministic solution of the equivalent difference equation

Measurements result in average concentrations  $a_i$  during the  $i$ -th time interval  $\{t_i, t_i+d\}$  of duration  $d$  [h], therefore also average values  $A_i$  and  $k_i$  can be only obtained from difference equations equivalent to the differential equation:

$$a_i' = -k_i a_i + A_i/V,$$

where the average differences  $a_i'$  have to be calculated by a numerical calcul from the measured  $a_i$ , or better from smoothed values. To calculate two unknown values  $k_i$  and  $A_i$  one needs to evaluate at least two adjacent intervals, but assuming the same values  $k$  and  $A$  for both intervals forces to fit a convex exponential course to experimental values also for concave courses resulting in meaningless negative exchange rates. One possible approach is to assume linear changes for both quantities  $k_i = k + \Delta k_i$  and  $A_i = A + \Delta A_i$  decreasing or increasing in accordance to the sign of  $\Delta k_i$  and  $\Delta A_i$ . Than four equations for four adjacent intervals have to be used for a general evaluation. Three equations can be used if the change of one of the quantity can be neglected and two equations are enough if  $A$  and  $k$  are nearly the same for two adjacent intervals.

### **Statistical solution of the equivalent difference equation**

An important disadvantage of the deterministic solution is the "uncompromising" fit of the solution to the input data  $a_i$  and  $a_i^*$  giving manytimes completely meaningless results or that it does not give uncertainties for the calculated results. This can be overwhelmed by a linear regression analysis using the difference equation as the statistical model with index  $i$  as the independent variable. This approach gives the covariance matrix for the four dependant results  $A$ ,  $k$ , and  $\lambda$ , of course the price for this is a larger number of intervals used for the evaluation with the same tendency of linear change of the entry rate and exchange rate. The optimal number of intervals suitable for evaluation can be determined by computer using the covariance matrix. Defining the difference equations symmetrically over the used intervals and shifting automatically the evaluation to the next interval the evaluation should give reasonable results.

### **Verification of the approach**

The evaluation method should be verified experimentally by independently measured time courses of the indoor radon concentration, of the radon entry rate and of the air exchange rate. This was up to now not practicable in our laboratories. Beside of verification by calculated time courses only evaluation of real time courses of indoor radon concentrations could be done. Unfortunately the results are not satisfactory entirely and the reasons of some failure are not completely understood. The lack of success for the deterministic approach were commented above. If the evaluated intervals of the time course contains a maximum or minimum the assumption of linearly changing rates are unrealistic of course but can be changed to more adequate ones. But the statistical approach results often in unrealistic high rates for the entry as well as the exchange but with the right ratio  $A/k$ .

### **Comment to the applicability of the compartmental model**

The compartmental or linear kinetic model used for the description of the indoor radon concentration assumes a perfect and immediate mixing of the entered radon in the whole volume of the room. This is of course not true and it has to be considered if this is not the reason for failures in the above described evaluation. It seems so that normal circulation of the indoor air in occupied rooms produced by heating systems during heating seasons but even by the entry of daylight through windows or by the inhomogeneity of insulation and temperatures of walls, floors and ceilings leads to quick and homogeneous enough mixing of radon entering mostly at the floor level. At least there may be a delay between the real entry time and the calculated effective entry of radon which could be corrected for.

### **Conclusion**

Two approaches are given for the determination of variable in time radon entry rates and air exchange rates from continuously measured indoor radon concentrations - numerical solution of the equivalent difference equations in deterministic or statistic form. The approaches are not always successful. Failures giving a right ratio for the searched rates but not of the rates themselves could not be explained.



## USE OF SSNTD IN THE CZECH REPUBLIC

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In the Czech Republic the legal limit for EEC is stated by Decree of Ministry of Public Health. Therefore the tremendous number of measurements is needed. In eighties the method for screening was searched and the recently only solid state nuclear track detector (SSNTD) is thought as cheap and effective approach.

In last five years more than 200 000 bare SSNTD were distributed in dwellings etc. Unfortunately about 30% is not coming back for EEC estimation (they are lost). The distribution of EEC results is possible to see in Appendix.

The journey of detectors from our Institute to dwelling was in 90% per cent through former District Hygiene Service Stations (departments of Radiation Protection). The chain process leading to inhabitant continued by mayors, local hygiene service stations, building offices etc. There are exceptions - there are distributing organisations and inhabitant could contact us directly (recent price of this service is more than 100 crowns).

In summer and autumn 1995 the new ways were organised as a result of radiation protection system changes. But the distribution of SSNTD was not stopped.

The response of bare detectors depends on radioactive equilibrium between radon and its progenies. The accuracy of measurement could not be better than 20% (with probability of 95 per cent uncertainty is less than 40%).

This fact pushed us to develop another system (RAMARN) - SSNTD in diffusion chamber. This kind is more frequent abroad, mostly as a result of radon gas investigation limit stated in most countries. In relevant region of concentration (more than 100 Bq.m<sup>-3</sup>) this system could be characterized by twice less "error". In future we try to popularise this system (SSNTD in diffusion chamber), but it has some disadvantages, e.g. dimension, the radon concentration is estimated (the limit of EEC being stated). The advantage is physical correspondence - the track density is equal to radon concentration time integral. (in case of bare SSNTD this correspondence does not exist).

Continual quality assurance is realized: For every thousand of detector 6 control detectors and 6 calibration detectors are etched simultaneously. The chief problem is influence of sunshine (the detectors are paler than calibration detector and the research of this influence has been started). Possibility of overestimation one could not exclude. Calibration irradiation is realized at radioactive disequilibrium so that wanted medium overestimation is reached (it corresponds to common conservative principle of radiation protection).

Other part of quality control is participation in international intercalibration. Unfortunately all these intercalibration is aimed to very low exposures (similar to background). It is caused by very short period of measurement in plenty countries (less than one year exposure typical for the Czech Republic). So that we thought results e.g. last NRPB comparison are not too important.

For the calibration of SSNTD the radon chamber (of State Metrologic Center - SMS) is used. The SMS methods of measurement is compared to the famous laboratories - EML New York, NPL Teddington, NRPB Chilton.

SSNTD are used also for special purposes - as <sup>210</sup>Po-<sup>210</sup>Pb detectors, to quantify the deposition process (wall effect).

In the Czech Republic there are three other SSNTD system applied:

1. ALTRAC Berlin detectors for one-week measurement
2. active personnel dosimeter OD88 (Czech made)
3. French active personnel dosimeter ALGADE

### Appendix

Summary of distribution of EEC (Bq.m<sup>3</sup>)

|       | <50   | 50-100 | 100-150 | 150-200 | >200   | total   |
|-------|-------|--------|---------|---------|--------|---------|
| 1991  | 5194  | 3999   | 1269    | 440     | 699    | 11601   |
| 1992  | 10593 | 7108   | 2633    | 1264    | 2883   | 24481   |
| 1993  | 16958 | 9366   | 3870    | 1781    | 2917   | 34892   |
| 1994  | 18722 | 16196  | 9293    | 3010    | 6307   | 53528   |
| total | 51467 | 36669  | 17065   | 6495    | 12 806 | 124 502 |
| \$    | 51351 | 36317  | 16689   | 6160    | 10 922 | 121 440 |

\$ special cases excluded

Special cases (1992-1993)

|          | <50 | 50-100 | 100-150 | 150-200 | >200 | total |
|----------|-----|--------|---------|---------|------|-------|
| Pluton   | 108 | 338    | 360     | 327     | 1821 | 2952  |
| Jáchymov | 8   | 14     | 16      | 8       | 62   | 108   |
| Starts   | 124 | 31     | 25      | 18      | 23   | 304   |

Starts are houses with elevated <sup>226</sup>Ra content,

Pluton is area in Central Bohemia with higher <sup>222</sup>Rn concentration in soil.

**Remark:** The distribution is deformed to higher values by using in nurseries and kindergardens (the system was originally designed for inhabited houses).



## TRACK ETCH DETECTORS WITH AIR GAP FOR MEASUREMENT OF RADON IN SOIL

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### Introduction

The applications of passive track etch detectors (TEDs) for the detection of alpha particles from Rn and its daughters has been commonly and successfully used for many years. The detector response (track density) is in general the sum of several components due to alpha particles from the Rn itself, from daughter products *plated-out* on the detector surface, on the walls of close objects and from daughter products deposited on airborne aerosols and dust particles. The usual method of Rn measurement in the soil gas is to place the plastic TEDs inside cups and bury them in drilled holes at several tens of cms below the surface of the soil. The contributions of the component mentioned above depend in this case mainly on the geometric arrangement of detectors in the cup and the cup volume.

Despite of good results of measurements, the method using the plastic TEDs inside cup has some principal disadvantages in routine application following from the necessity of the cup itself. Its volume (200-250 ml as a rule) determines the diameter of the drilled hole and consequently the final costs. It is necessary to consider also the consumption of metalized plastic foils for hermetic sealing before and after measuring period and sometimes even mailing costs. Plastic cups are also relatively fragile considering the usual field conditions.

The main aim of this study was to develop a method of Rn concentration measurements in soil using TEDs without cups. Our approach enables to minimize the detector dimensions resulting into smaller diameter of drilled holes, more rigid construction, easier handling and mailing, lower consumption of material and consequently in lower costs.

### Principle, Arrangement And Treatment Of Detectors

We have utilised our previous experience with the application of the electrochemically etched CR-39 TED for detection of alpha particles. To avoid the detection of alpha particles in the broad energy spectrum (0-8.8 MeV in case of bare detector and <sup>222</sup>Rn, <sup>220</sup>Rn and their daughters) the detectors were arranged into parallel pairs with the active (detecting) surface faced opposite one to another and with the narrow air gap between them. Two small metallic holders glued together using adhesive tape keep the detectors parallel in a properly chosen distance **h**. This arrangement has the following purposes:

- a) define the rectangular air volume from which the alpha particles can be registered
- b) enable to control the detector response by changing **h**
- c) reduce the *plate-out* effect from outside (the surface of side opening is relatively small with respect to detector surface)
- d) create thermally and electrically well conductive capsule reducing humidity condensation and electrostatic effects

For the above arrangement the theoretical number **N** of alpha particles capable create tracks in the volume **V** of air in the gap between the detectors can be expressed as

$$N = VA_t\sigma \quad (1)$$

where  $A_v$  is volume activity,  $t$  exposure time and  $\sigma$  registration efficiency. Because of  $V = Sh$  ( $S$  is the detector surface) and  $\sigma = 1 - \sin\phi_{cr}$ , where  $\phi_{cr}$  is the critical angle of incidence, the corresponding track density  $D$  on each detector of the pair can be obtained from (1) as

$$D = 0.5Aht(1 - \sin\phi_{cr}) \quad (2)$$

The constant 0.5 conclude from  $2\pi$  geometry of irradiation. The value of the critical angle of incidence  $\phi_{cr}$  depends on the particle energy and the etching conditions but it can be estimated from our previous measurements. Each alpha particle emitted inside the gap and registered in the form of etched track has the energy in the interval  $(E_0 - \delta E, E_0)$ , where  $E_0$  is the primary energy and  $\delta E$  is the energy loss along its trajectory in the air between the point of its origine and the impact point at the detector surface. Considering the real arrangement ( $h < 5$  mm), the length of the trajectory in the air is only few milimeters and therefore  $\delta E$  does not overcome several hundreds keV. In the first approximation alpha particles can be considered as the monoenergetic ones and the experimental value of  $\phi_{cr} = 30^\circ$  for  $E = 5.5$  MeV (valid for etching conditions described below) can be used and Eq.2 can be simplified into the form

$$D = 0.25A_vht \quad (3)$$

Assuming  $h = 1$  mm,  $A_v = 100$  kBqm<sup>-3</sup> and  $t = 1$  week, we can estimate the lower limit of corresponding track density by value  $\sim 1500$  tracks cm<sup>-2</sup>, i.e.  $\sim 90$  tracks cm<sup>-2</sup> per 1 MBqm<sup>-3</sup>h. In the real case these values should be  $\sim 2-3$  times higher due to contribution of alpha particles from daughter products. Because of  $h$  can vary within 0.5 - 5 mm, this arrangement provides enough possibilities for optimization of the measurement either with respect to exposure time or the expected volume activity.

As detectors the square sheets 14x14 mm<sup>2</sup> of the CR-39 material (Pershore Mouldings, standard grade, 32 hours cure, 500  $\mu$ m) were used. The detectors were treated by combined chemical and electrochemical etching in 30 % KOH (density 1.29 gcm<sup>-3</sup>). Chemical etching at 70°C takes 2 hours (removed layer  $\sim 6$   $\mu$ m), the electrochemical etching in the same solution at 25°C, 4 hours at 10 kHz and 25 kV<sub>peak</sub>cm<sup>-1</sup>.

### Calibration

Several sets of detectors of different air gap width were exposed to alpha particles from Rn in soil at two experimental study sites in Stráž pod Ralskem, Northern Bohemia. The first one is the reference site where a soil radon concentration is known and it was measured formerly during the 14-month observation. The second one is situated on the old part of a uranium mill tailings. Our detectors were hanged in the permanently inserted metallic tube of inner diameter 20 mm at 80 cms below the surface. Exposure times varried between 3 and 11 days. The actual soil radon concentrations were measured using Lucas cells either daily or at least 6 times within the exposure time. The calculated integral soil radon concentration value varied between 2 and 38 MBqm<sup>-3</sup>h. The response as a function of air gap width is given in Tab.1. The value of response for  $h = 1$  mm lies really with- in the estimated interval 180-270 tracks cm<sup>-2</sup> per 1 MBqm<sup>-3</sup>h.

In order to prove a linearity of the response a serie of relative measurement was carried out in the same experimental arrangement as in the previous case, but in the garden of Dept. of Radiation Dosimetry. The simultaneous exposures of detectors with different air gap width were carried out several times in period of 1994-5 and their responses were compared to the reference

width 1 mm. From the results of measurements concludes that response of detectors in the parallel arrangement is a linear function of  $h$  up to  $\sim 4$  mm.

**Tab. 1** Response (in trackscm<sup>-2</sup> per MBqm<sup>-3</sup>h) as a function of air gap width  $h$

|                     |        |        |         |        |        |
|---------------------|--------|--------|---------|--------|--------|
| Gap width [mm]      | 0.5    | 1.0    | 1.3     | 1.5    | 2.0    |
| Response to soil Rn | 117±15 | 220±27 | 280±117 | 365±54 | 395±28 |

Considering as the limits of measurable track density the usual background value ( $\sim 50$  cm<sup>-2</sup>) and the max. evaluable track density ( $\sim 10^4$  cm<sup>-2</sup>) the practical limits for application of the detector described can be calculated. For the case of  $h = 1$  mm these limits are summarized in Tab.2.

**Tab 2** Practical limits for application (gap width 1 mm)

| Rn conc. [Bq/m <sup>3</sup> ] | Applicable exposure time | Exposure time | Measurable Rn concent. [Bq/m <sup>3</sup> ] |
|-------------------------------|--------------------------|---------------|---|
| 1k                            | 9days - 5years           | 1day          | 9k - 1.9M                                   |
| 10k                           | 1day - 6month            | 1week         | 1.4k - 270k                                 |
| 100k                          | 2hours - 19days          | 1month        | 320 - 63k                                   |
| 1M                            | 15minutes - 2days        | 1year         | 26 - 5.2k                                   |

### Conclusions

The parallel arrangement of two track etch detectors in the open metallic holder seems to be promising as the complementary method to the commonly used *cup-technique* for radon measurement. The firmness, simple and compact construction, small size as well as low costs could be successfully utilized mainly in field measurements. The possibility of a variable sensitivity by the distance between the detectors makes the system versatile for many applications, 2 detectors with different  $h$  can practically exclude the case of under- or overexposure. The more precise calibrations including exposures in Rn-chambre and study of an eventual influence of humidity are supposed to be done in the nearest future.



## RADON IN GEOLOGICAL MEDIUM

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### Introduction

The distribution of a Rn-222 in subsurface layer of the geological medium is current task of the environmental projects in the regions of city agglomerations in Slovakia.

For example, in frame of the Bratislava-environment, abiotic component project (1990-1993) and Košice-abiotic component of the environment project (1994-1999?), the compilation of radon risk maps is among other subprojects.

The paper presented deals with behaviour of the radon in geological medium and with some results of the radon survey in Bratislava and Košice regions.

### Generally

The Rn-222 very easy penetrates through permeable rock complexes and active faults to the distances up to first kilometers from the source. The content of the radon in soil air depends on Ra-226 content in the rocks, emanation ability of minerals and rocks, permeability of rock formations for water and gas.

In the rocks and neotectonics, the radon spreads by diffusion and convection. The diffusion movement is affected by physical properties of the geological medium, while convection flow originates in consequence of physical conditions changes, mainly temperature and pressure. The transport of radon by convection is higher than by diffusion.

The distribution of radon and values of its volume activity ( $a_v$ ) in subsurface layer is influenced by climatic changes. The radon rising up from soil air to the surface increases with increased air temperature and decreases with high air pressure, humidity of atmosphere and rainfall. In dry period the  $a_v$  values in soil are low, and opposite.

The risk of radon penetrating from the subsurface layer to the houses depends on  $a_v$  value in the soil air and on structural-mechanic properties of the basement soils.

The assesment of the radon risk in Bratislava and Košice regions is based on methodology in Czech Republik and on Notice of Ministry of health of the Slovak Republic No.406/92. The assesment of the soil gas-permeability is in concordance with previous Czechoslovak standard No. 73 1001 (See Table below).

| Radon risk category | Volume radon activity [kBq.m <sup>-3</sup> ] |         |         |
|---------------------|--|---------|---------|
|                     | soil permeability                            |         |         |
|                     | low  | medium  | good    |
| low - I             | < 30   | <20     | <10     |
| medium - II         | 30 - 100                                     | 20 - 70 | 10 - 30 |
| high - III          | >100   | >70     | >30     |

### Radon Risk Maps

#### 1. Bratislava region

The  $a_v$  has been detected in the holes of 0.80 m deep. The density of observations - 3 reference areas (one represents 20 stations) per 1 sq.km. The radon risk maps in 1:25 000 and 1:50 000 scales have been compiled. The 56.8% of the project area lies in low radon risk, 37.6%

in medium radon risk and 5.6% in high radon risk (See Fig.1). Follow-up monitoring of the equivalent volume radon activity (EVRA) at the flats, located in the areas with high radon risk of the surface layer, has showed values several times higher than Slovak limits (Marianka, Rača, Vajnory).

The evidence that neotectonics is excellent medium for rising up radon emanation to the subsurface layer, is shown in Fig.2. The tectonic zone of Líščie údolie in Bratislava-Karlova Ves area has been clearly detected by profile radon survey.

## 2. Košice region

At present, northern half of the area in question was covered by radon survey. The low and medium radon risks have been observed here, while localities with high radon risk are small in extent.

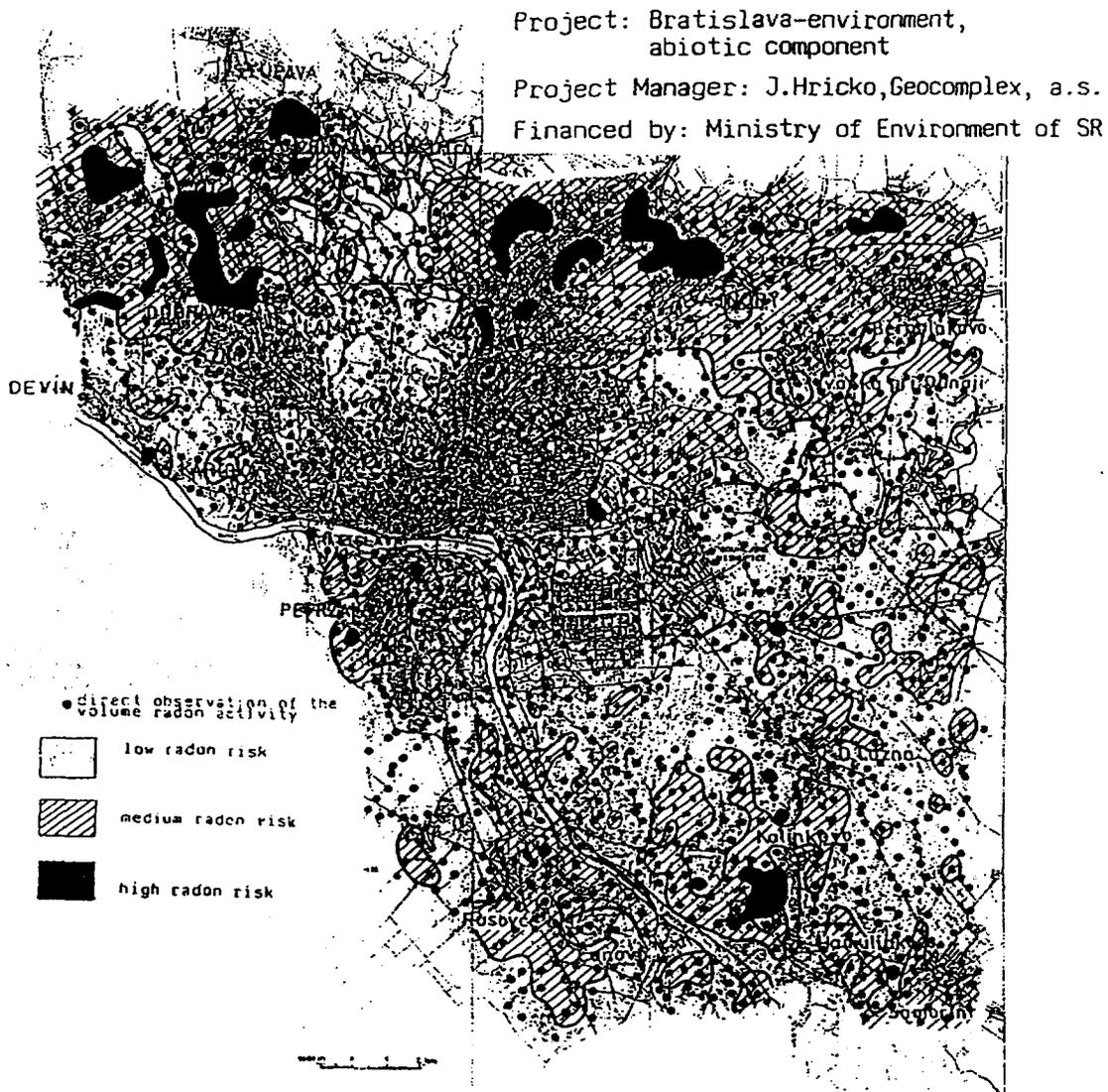
The part of radon risk and soil permeability map from northern Košice area is shown in Fig.3.

## Acknowledgements

We wish to thank Dr.K.Együd and Dr.L.Andor from Ministry of Environment of the Slovak Republic who allowed authors of this paper to use the results of Bratislava and Košice environmental projects. We wish also to thank other authors of the maps used in this paper.

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- Hricko J., 1995, Košice-abiotic component of environment: Progress report, Manuscript, Geocomplex a.s., Bratislava.



| Radon risk category | Volume radon ( <sup>222</sup> Rn) activity /kBq.m <sup>-3</sup> / Soil permeability |        |       |
|---------------------|---|--------|-------|
|                     | low   | medium | good  |
| low - I             | <30   | <20    | <10   |
| medium - II         | 30-100  | 20-70  | 10-30 |
| high - III          | >100  | >70    | >30   |

Fig.1. Radon risk map of the Great Bratislava region  
(by P.Čížek, H.Smolárová 1992, J.Hricko 1993)

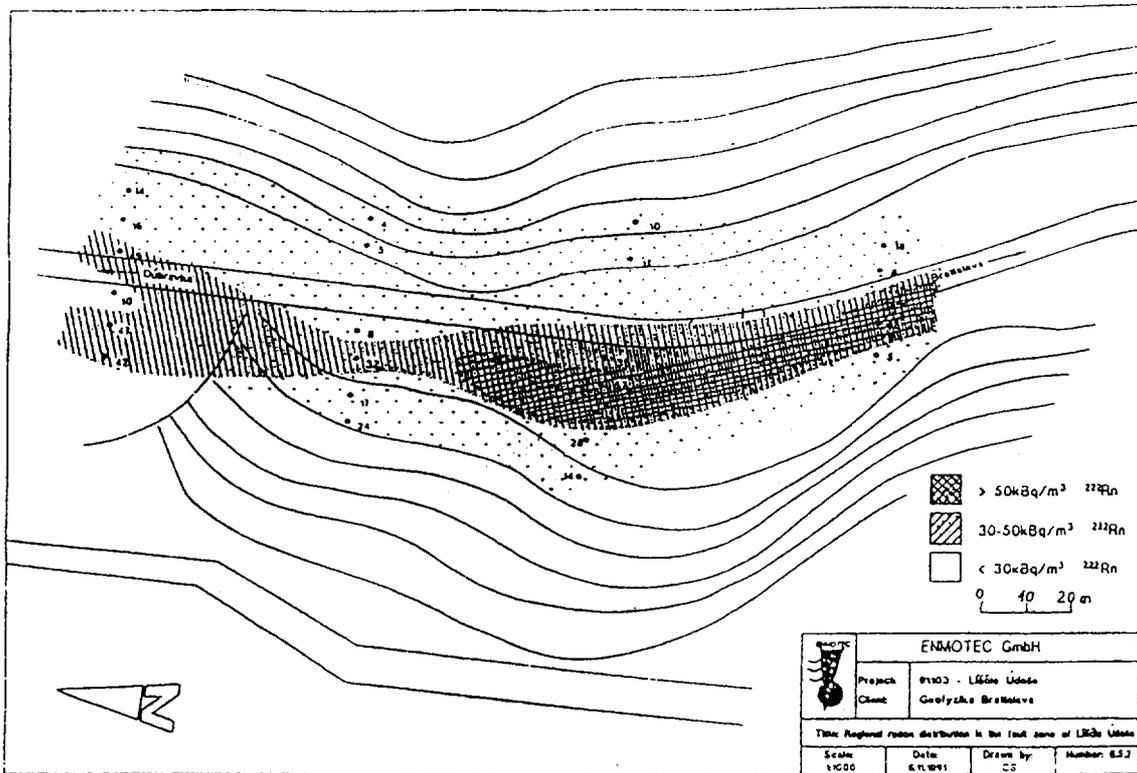


Fig.2. Regional radon distribution in the fault zone of Líščie údolie valey (Bratislava - Karlova Ves)

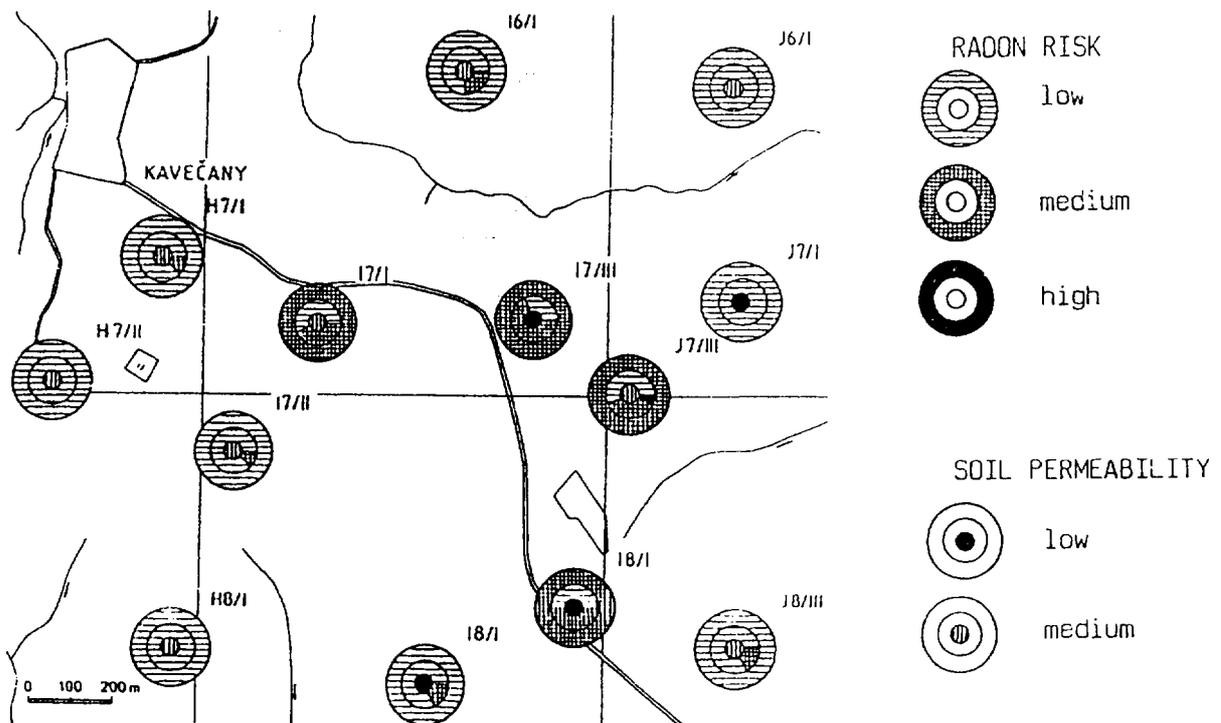


Fig.3. Radon risk map from northern part of Košice region (by F. Suchý, S. Daniel 1995)



## VARIATIONS OF $^{222}\text{Rn}$ CONCENTRATION IN OUTDOOR ATMOSPHERE AND IN SOIL AIR

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### Introduction

$^{222}\text{Rn}$  is produced by alpha decay of  $^{226}\text{Ra}$  in soil. A small fraction of totally produced  $^{222}\text{Rn}$  escapes from soil particles into soil air. Then  $^{222}\text{Rn}$  is transported predominantly by molecular diffusion to outdoor atmosphere.

In the soil air the  $^{222}\text{Rn}$  concentration reaches values of several  $\text{kBq}\cdot\text{m}^{-3}$ . In the outdoor atmosphere, a radon concentration is thousand times lower. The radon concentration in the soil air as well as in the outdoor atmosphere is not stable. It varies irregularly depending on meteorological conditions. However, there were found out regular daily and seasonal variations of  $^{222}\text{Rn}$  concentration in both medium [1-3]. Mainly the variations of  $^{222}\text{Rn}$  concentration in soil air have not yet been studied sufficiently up to now.

Some more significant results of our study of  $^{222}\text{Rn}$  variations in outdoor atmosphere as well as in soil air are shown in this report.

### Methods

The  $^{222}\text{Rn}$  concentration in the outdoor atmosphere has been studied at our department since 1987. Since the November 1993 we have also measured the  $^{222}\text{Rn}$  concentration in the soil air.

Up until the end of 1991 y the method of measurement of radon concentration in outdoor atmosphere was as follows. Radon was concentrated from the air volume of 10 l on an active carbon and after its transfer radon was measured by means of the scintillation chamber of Lucas-type [4]. A sampling was realized every morning at 9 o'clock in the height of 1,5 m above the ground surface. Since Februar of 1991, radon in the outdoor atmosphere has been monitored continously by a large volume scintillation chamber which volume is 4,5 l [4]. The continual measurements of the radon concentration in the soil air have been realized in the same way by the use of the scintillation chamber of Lucas-type which volume is 125 ml.

### Results

#### 1. $^{222}\text{Rn}$ concentration in outdoor air.

By continual measurements of  $^{222}\text{Rn}$  concentration in the outdoor atmosphere of Bratislava there were obtained about 80.000 data of  $^{222}\text{Rn}$  concentration. The results arranged in this way make possible to demonstrate the average daily courses of  $^{222}\text{Rn}$  concentration for individual months and average annual courses for various time intervals.

The average daily course of  $^{222}\text{Rn}$  concentration calculated on the basis of all measurements has wave form with maximum between 4 and 6 a.m and with minimum between 2 and 4 p.m. The  $^{222}\text{Rn}$  concentration reaches its average daily value at 9.30 a.m and at 9 p.m. in the evening. The ratio of the maximum and minimum radon concentrations is equal to 1.7.

The average annual courses of  $^{222}\text{Rn}$  concentrations calculated on the basis of continual measurements during the years 1991-94 for various time intervals are shown in Fig.1. The average annual course reaches the maximum values in the autumn months and the minimum values in the end of spring till beginning of summer. Further we can see, that the average annual course of  $^{222}\text{Rn}$  concentration calculated from all the data is in good agreement with the average annual

course of  $^{222}\text{Rn}$  concentration calculated on the basis of measurements made between 8 and 10 p.m.

The above mentioned results serve mainly for a verification of models describing the radon variations in the outdoor atmosphere and for an improvement of their accuracy [5].

## 2. $^{222}\text{Rn}$ concentrations in soil air.

At the continual measurement of  $^{222}\text{Rn}$  concentration in a soil air, the air was collected out of the depth of 0.8 m below the ground surface. The sampling place of the soil air was situated near the position at which an air also was taken for the determination of  $^{222}\text{Rn}$  concentration in outdoor atmosphere. The soil of this place is fine grained. The content of fine particles with diameters of  $d < 60 \cdot 10^{-6} \text{ m}$  was about 68 %. The further soil characteristics were as follows:  $A_{\text{Ra-226}} = 37,5 \text{ Bq} \cdot \text{kg}^{-1}$ , the total porosity = 0.55, the dry bulk density =  $1090 \text{ kg} \cdot \text{m}^{-3}$ , emanation coefficient = 14,5 % at the volumetric soil moisture content 19 %.

The saturated concentration of  $^{222}\text{Rn}$  calculated on the basis of mentioned parameters is equal to ( 16 360 1810 )  $\text{Bq} \cdot \text{m}^{-3}$ . This value is reached in the depth about 2 m.

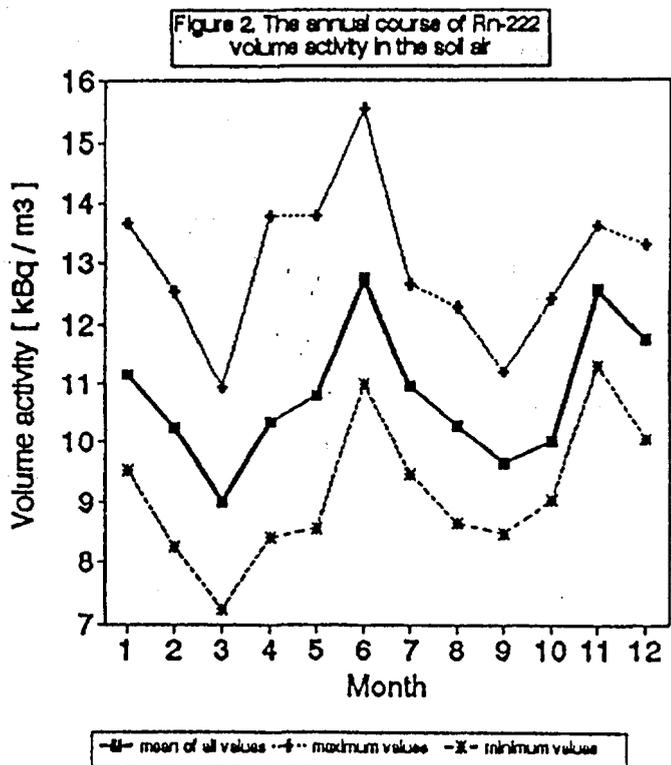
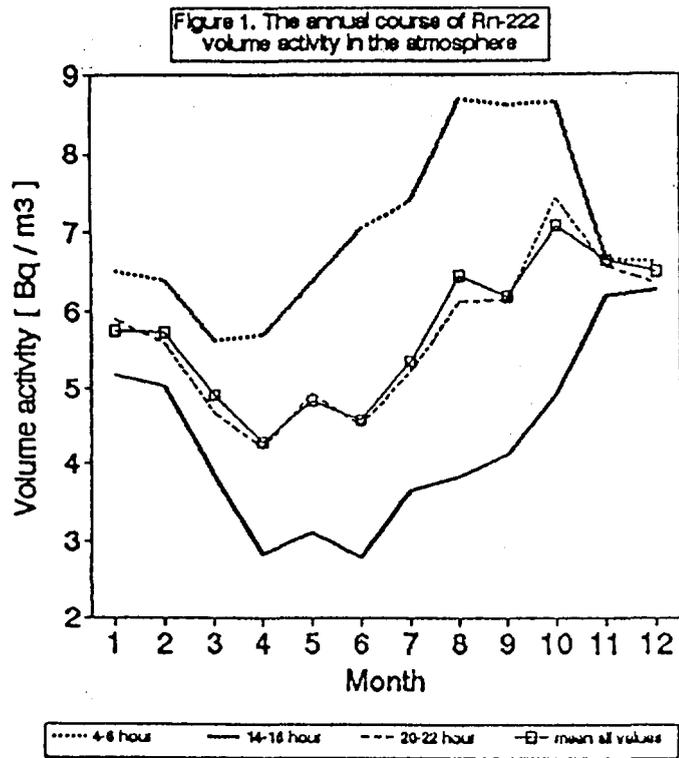
The average annual course of  $^{222}\text{Rn}$  concentration in a soil air is given in Fig.2. There are also shown the measured maximum and minimum values of  $^{222}\text{Rn}$  concentration in the course of individual months. In April and May, the measured range of  $^{222}\text{Rn}$  concentrations with regard to the average annual value is about 50 %. The increase of  $^{222}\text{Rn}$  concentration in a soil air during a spring and in the beginning of summer is connected obviously with rainfall in this season. This similar annual courses were also published in [2,6].

Also daily variations of  $^{222}\text{Rn}$  concentration in soil air were found out. The amplitude of these variations reaches maximum only 4 % of an average monthly value of  $^{222}\text{Rn}$  concentration. However, these results allow to study an influence of variations of atmospheric pressure on  $^{222}\text{Rn}$  concentration in soil air.

From practical point of view, the obtained results of measurements of  $^{222}\text{Rn}$  concentration in soil air suggest, that a representative season for realization of radon measurements in the field could be months from July to October. In these months not only the minimum dispersion of measured  $^{222}\text{Rn}$  concentrations was obtained, but also the average monthly values of  $^{222}\text{Rn}$  concentration were very close to the annual average of  $^{222}\text{Rn}$  concentration in a soil air.

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## SHIELDING OF GAMMA FIELD IN RESIDENTIAL HOUSES

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### Summary

In the past some flats were built from defective materials contained <sup>238</sup>U, which radiate dangerous gamma radiation. The object of this work consisted in searching mechanical barriers, which would decrease penetrating of this radiation into flat.

The measurement was realized in system made of connecting of Ge/Li detector with multichannel analyser MCA JAK 202 and IBM PC.

This measurement was carried out without shading Ge/Li detector by Pb, which makes certain and unnegible contribution by reflection and dispersion of radiation to the whole detection yield.

Plenty of building parts such as bricks, plaster slabs with/without Pb dust, wasted plaster from Počerady Electric Power Station, etc. were measured to get and compare shading abilities.

Maximal intensity of gamma radiation (47.1 %) is visible for energy  $E = 609 \text{ keV}$  <sup>226</sup>Ra, therefore the measurement was only carried out for this energy.

The measurement performed in defective houses START during years 1988 - 1991 demonstrated that excepting higher activity <sup>222</sup>Rn and its daughter products forms unnegible gamma field, as well. This one is limited by values of rate dose equivalent. The screening of gamma field in flats and houses was a part of The State Target N - 03 - 326 - 830. The problem was succesfully solved by lead slabs fixed to wood construction that is covered by applications. This problem was solved in years 1993 - 94 again <sup>1)</sup>. The manipulation with materials and construction was difficult, therefore another materials and segments were tested, for more easy fix to defective walls. In 1995 the experiment was realised in the cooperation with the chemical department of Počerady Electric Power Station, the plaster is outlet product from the removing sulphur process. There were made an experimental slabs, sizes 18 x 18 x 2 cm. The barrier effect of slabs were compared with other building material and parts.

### Results of Measuring

The results of measuring are summared in TAB.I. There are present values of linear absorption coefficient /  $\mu$ /, further the values of half-layer  $x_{1/2}$  and square weighting of 1 half-layer of inquired material ( $m^2$ ).

### Conclusion

So that the elimination of radiation would be effective is necessary reduce the level of radiation penetrating to the smallest level. However, the thickness of shading material is limited by economical reasons, prices of material, square weighting and reducing of living room.

The results of measuring is this one:

The plaster slabs with Pb dust made in EPS Počerady are suitable to reduce gamma ray, the values of reducing coefficient are very high in comparison with other samples and values of square weighting are low.

Tab. 1

| SAMPLE                              | LINEAR ABSORB. COEFFICIENT [m <sup>-1</sup> ] | HALF - LAYER [cm] | SQUARE WEIGHTING [kg.m <sup>-2</sup> ] |
|-------------------------------------|---|-------------------|--|
| plaster                             | 8,86  | 7,82              | 86,77                                  |
| plaster + sand (1:1) + 50 w % Pb    | 13,89   | 4,99              | 105,09                                 |
| plaster + sand (1:1) + 25 w % Pb    | 5,74  | 12,08             | 253,53                                 |
| plaster + sand (1:1) + 33,33 w % Pb | 13,77   | 5,03              | 114,1                                  |
| plaster (Počerany)                  | 10,43   | 6,65              | 95,89                                  |
| plaster (Počerany) + 50 w % Pb      | 18,67   | 3,71              | 74,42                                  |
| plaster (Počerany) + 66,67 w % Pb   | 20,98   | 3,31              | 80,19                                  |

#### Literature

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## HISTOLOGIC TYPES OF LUNG CANCER IN URANIUM MINERS

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### Introduction

It has been recognized that lung cancer risk in uranium miners is associated with increased incidence of certain histologic types, especially epidermoid and small cell type.

Recent results showed that the basic dependence of the relative risk for the two main histologic types (epidermoid and small cell) is linear with cumulative exposure. However, time and age modifiers of the dependence may be different for the two types.

The aim of the study was firstly to verify the assumed differences in incidence of histologic types of lung cancer for the studied cohort and general population and secondly to characterize the relation of histologic type specific incidence to different exposure patterns.

### Methods

The cohort study is based on data of the oldest Czechoslovak cohort. The cohort includes 4320 former uranium miners who started their work 1948-59 and worked at least 4 years. The cohort was divided in two groups according to exposure rate pattern in order to analyze the impact of exposure conditions on main histologic types incidence. In group 1 the exposure rate never exceeded the level of 5 WL since the third year of exposure, while in group 2 the exposure rate exceeded at least once the level of 5 WL. The first two years of exposure had not been taken into consideration in order to confirm the hypothesis about the inhibition effect of high exposure rate levels. Consequently, these could influence lung cancer risk caused by previous exposures.

In accordance to the WHO classification and from previous data analysis (Ševc et al., 1989), lung cancer cases were divided into four groups:

- 1 - epidermoid
- 2 - small cell
- 3 - adenocarcinoma
- 4 - other histologic types

### Results

A total of 705 lung cancer cases were recorded by 1990. Morphological diagnoses were available in 458 cases (65% histologic type specification), histologic types could not be specified in 41% cases (i.e. 287 cases), as shown in Table 1.

Table 2 summarizes type specific mortality for two main histologic types in dependence on attained age. The decrease of relative risk with age is apparent in both histologic types. A more detailed analysis shows that decrease of relative risk in the epidermoid type is less rapid than in the small cell type. Therefore the risk of small cell carcinoma will be significantly lower than the risk of epidermoid carcinoma in higher ages. This is the contrary of situation in younger age groups of miners.

Table 3 demonstrates dependence of relative risk on cumulated exposure (lagged by 5 years) for the two main histologic types. The relative risks show similar values in the corresponding exposure categories (by exposure is meant cumulated 5 year lagged exposure).

**Tab. 1** Distribution by histologic types

| Histologic type | Code | Cases | %    |
|-----------------|------|-------|------|
| no information  | 0    | 201   | 28,5 |
| epidermoid      | 1    | 173   | 24,4 |
| small cell      | 2    | 185   | 26,2 |
| adenocarcinoma  | 3    | 31    | 4,4  |
| other types     | 4    | 29    | 4,1  |
| unspecified     | 5    | 40    | 5,7  |
| no material     | 9    | 46    | 6,5  |
| Total           | -    | 705   | 100  |

**Tab. 2** Age and histologic type specific mortality

| Type  | Small cell |       | Epidermoid |       |
|-------|------------|-------|------------|-------|
|       | 0          | 0/E   | 0          | 0/E   |
| - 44  | 24         | 35,02 | 7          | 10,64 |
| 45-54 | 71         | 11,97 | 39         | 7,20  |
| 55-64 | 65         | 4,27  | 81         | 5,39  |
| 65-84 | 25         | 2,65  | 46         | 3,79  |
| Total | 185        | 5,91  | 173        | 5,2   |

**Tab. 3** The dependence of relative risk on cumulated exposure for main histologic types

| Type           | Small cell |       | Epidermoid |       |
|----------------|------------|-------|------------|-------|
|                | 0          | 0/E   | 0          | 0/E   |
| Exposure (WLM) | 0          | 0/E   | 0          | 0/E   |
| 0-99           | 18         | 2,72  | 17         | 2,42  |
| 100-199        | 69         | 5,03  | 62         | 4,26  |
| 200-299        | 48         | 8,36  | 42         | 6,98  |
| 300-399        | 21         | 7,72  | 21         | 7,07  |
| 400-           | 29         | 11,68 | 31         | 11,52 |

**Risk models**

The analysis of epidemiological studies showed that time since exposure and attained age considerably influence excess relative risk of lung cancer. In this study, the BEIR IV model (1988) has been applied in the following form:

$$RR(a) = 1 + b(w_1 + q_2 w_2 + q_3 w_3 + q_4 w_4) g(a),$$

where  $w_1, w_2, w_3, w_4$  are exposures experienced in periods 5-14, 15-24, 25-34, 35+ years previously,  $b(=ERR/WLM)$  is the excess relative risk per WLM,  $g(a)$  is age modifier.

We have studied influence of the basic variable - cumulated exposure and main factors modifying the exposure - effect relationship, namely:

**1) time since exposure (TSE)**

**2) age at exposure (AE)****3) time pattern (TP)**

The coefficient of ERR/WLM was 5 times higher in small cell than in epidermoid type. The effect of time pattern was found significant in the small cell type and not in the epidermoid type, which confirm the hypothesis about parallel inhibition effect of alpha radiation on small cell type of lung cancer. Significant negative values of the AE parameter confirm the theory about decrease of lung cancer risk with age at exposure. This trend is more apparent for the small cell type.

**Tab. 4** Relative risk model for histologic types

|         | Epidermoid | Small cell |
|---------|------------|------------|
| ERR/WLM | 0,0661     | 0,3286     |
| TSE     |            |            |
| 5-1     | 1,000      | 1,000      |
| 15-2    | 0,926      | 0,110      |
| 25-34   | 0,150      | 0,014      |
| 35-     | -0,016     | 0,008      |
| AM      | -0,0338    | -0,0878    |
| TP      | 0,708      | 0,534      |

**Summary**

The recent results confirm linear dependence of exposure and relative risk, decrease of alpha radiation effect with time since exposure and decrease of effect in older age categories.

A detailed analysis of exposure - effect relationship for the two main histologic types of lung cancer confirms hypothesis formulated by J. Ševc about parallel inhibition effect of alpha radiation in higher exposure rates which followed after period of initiation of malignant process of bronchial epithelium cells. This phenomenon was observed only in small cell type of lung cancer.

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## EXPERIMENTAL STUDY OF RADON AND THORON DIFFUSION THROUGH BARRIERS

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### Introduction

The measurement results of diffusion parameters for radon ( $^{222}\text{Rn}$ ) and thoron ( $^{220}\text{Rn}$ ) through barriers, experimental equipments and theoretical background of diffusion are presented in this paper. The diffusion barriers are used for measuring radon and thoron by passive detectors and like one of the reduction techniques in houses.

The choice of diffusion barrier is very important and it should have following properties:

- barrier for measuring and determination of radon and thoron activities must remove radon and thoron daughters, water vapour if possible and discriminate radon and thoron
- barrier for reduction of radon in a dwelling must lower radon and thoron gas fluxes from soil and building materials to room so that radon activity will be reduced under action level into it, good long life mechanical and chemical properties, without of containing radium and unhealthy materials.

The knowledge of basic parameters of barrier such as diffusion coefficient and porosity is important in prediction of radon and thoron transport through it. From this prediction it is possible to estimate the efficiency of passive dosimeters with barrier, their minimum time of exposure. The radon concentration in room for given barrier in remedial action will also be estimated.

### Materials and Methods

The measuring equipment for estimation of diffusion coefficient for radon was built according to general diffusion theory where radon transport is from volume  $V_1$  (radon chamber) to volume  $V_2$  (diffusion room) through diffusion barrier. This equipment is shown in Fig.1 and it was described in work [1]. Constant radon activity in radon chamber is controlled by radon source. Radon activities are measured continuously by radon monitor and by Lucas scintillation cells sampling one or two times per day in chamber and diffusion room. Exposure time for determination of the diffusion coefficient of barrier is from one day to one week in dependence on properties of barrier.

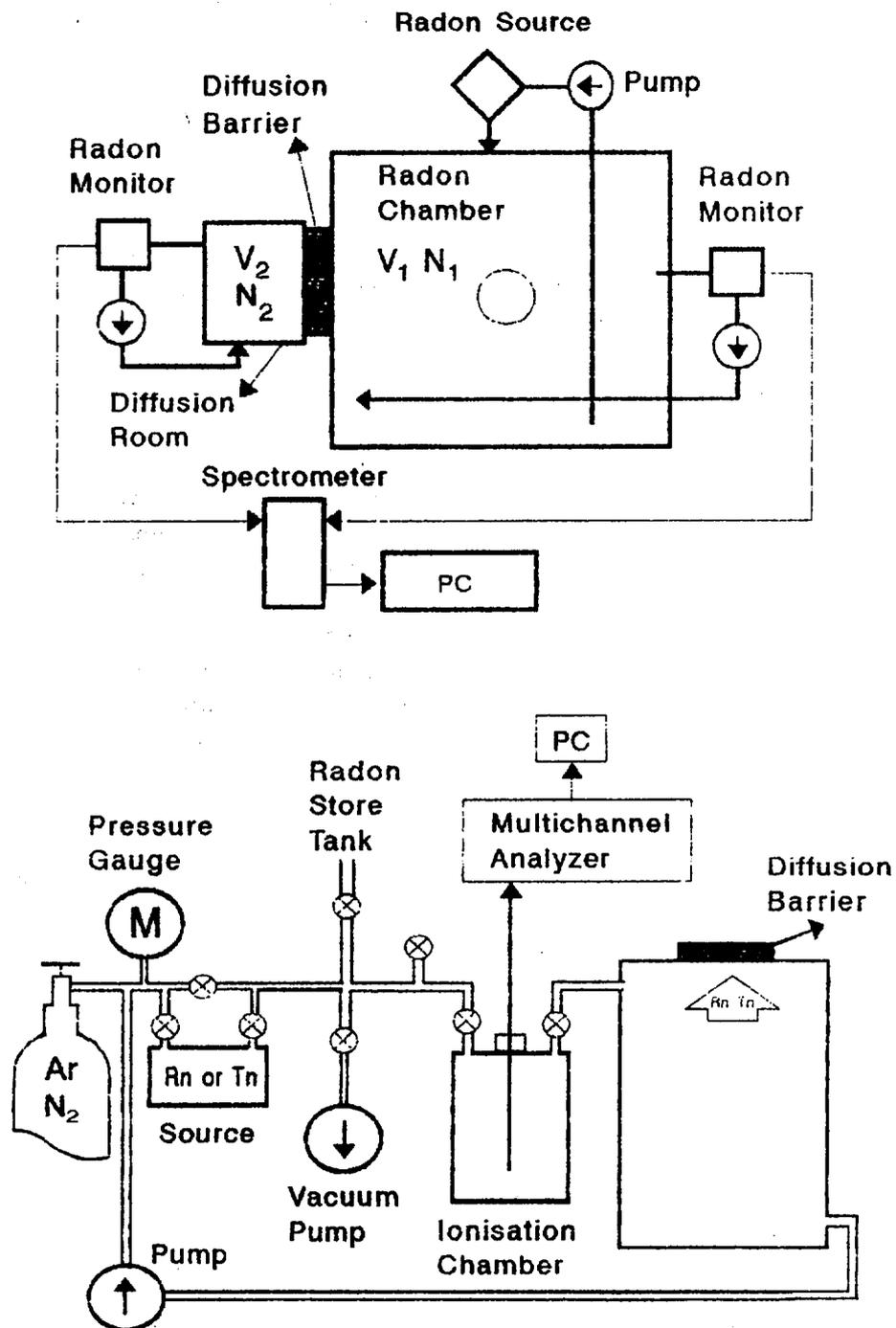
The diffusion of thoron and radon was studied by further measuring apparatus which is schematic drawn on Fig.1. Thoron or radon gas flew from exposure chamber through barrier to free air of room. Thoron or radon gas is added continuously to chamber from the source. Activities are calculated from spectra measured by ionisation chamber filled with nitrogen like working gas.

The pressure of air, is measured on the both sides of barrier, is the same.

### Theory

The volume activities  $A_1$  and  $A_2$  of radon or thoron for geometry sketched in Fig.1 are described by kinetic equations for the gas diffusion process as follows [2]

$$\frac{dA_i}{dt} = -\lambda A_i + (2-i)Q_i + \frac{\phi S}{V_i} + (-1)^i (A_1 - A_2) \frac{KS}{V_i h}, \quad i=1,2. \quad (1)$$



**Fig. 1** Schemes of measuring arrangement for the determination of radon and thoron diffusion parameters.

The symbols in equations represent: radon decay constant  $\lambda$ , radon production rate  $Q$ , radon exhalation rate from barrier  $\phi$ , permeability  $K$ , thickness of barrier  $h$ , diffusion area  $S$ .

The permeability of material to gas can be defined by equation

$$K=SD \tag{2}$$

where  $S$  is solubility and  $D$  is diffusion ability of the gas in the barrier. The value of solubility for simple gases like radon or thoron is close to one in polymers and therefore  $K=D$  [3].

Going out from the experimental possibilities, there are various solutions of equations

(1). In our paper are presented solutions for three boundary conditions, as follows:

a) The solution for constant radon activity  $N_1$ , initial activity  $A_0$  in volume  $V_2$  and  $\phi=0$  is given by

$$A_2(t) = \frac{DSA_1}{hV_2L}(1 - e^{-\lambda t}) + A_0e^{-\lambda t} \tag{3}$$

where

$$L = \lambda + \frac{DS}{hV_2} \tag{4}$$

b) If radon activity  $A_{11}$  is added to volume  $V_1$  at the beginning of measurement,  $\phi = 0$  and  $A_0$  is the initial activity in volume  $V_2$ . The solution of the equations (1) is given by

$$A_2(t) = A_{11}(e^{-\lambda t} - e^{-\lambda t}) + A_0e^{-\lambda t} \tag{5}$$

c) The solution of the equations (1) is derived from a second equipment, where radon diffuses from volume  $V_1$  to free air in a room is given by

$$A_1(t) = \left(\frac{Q}{L} + \frac{DSA_2}{hV_1L}\right)(1 - e^{-\lambda t}) + A_{01}e^{-\lambda t} \tag{6}$$

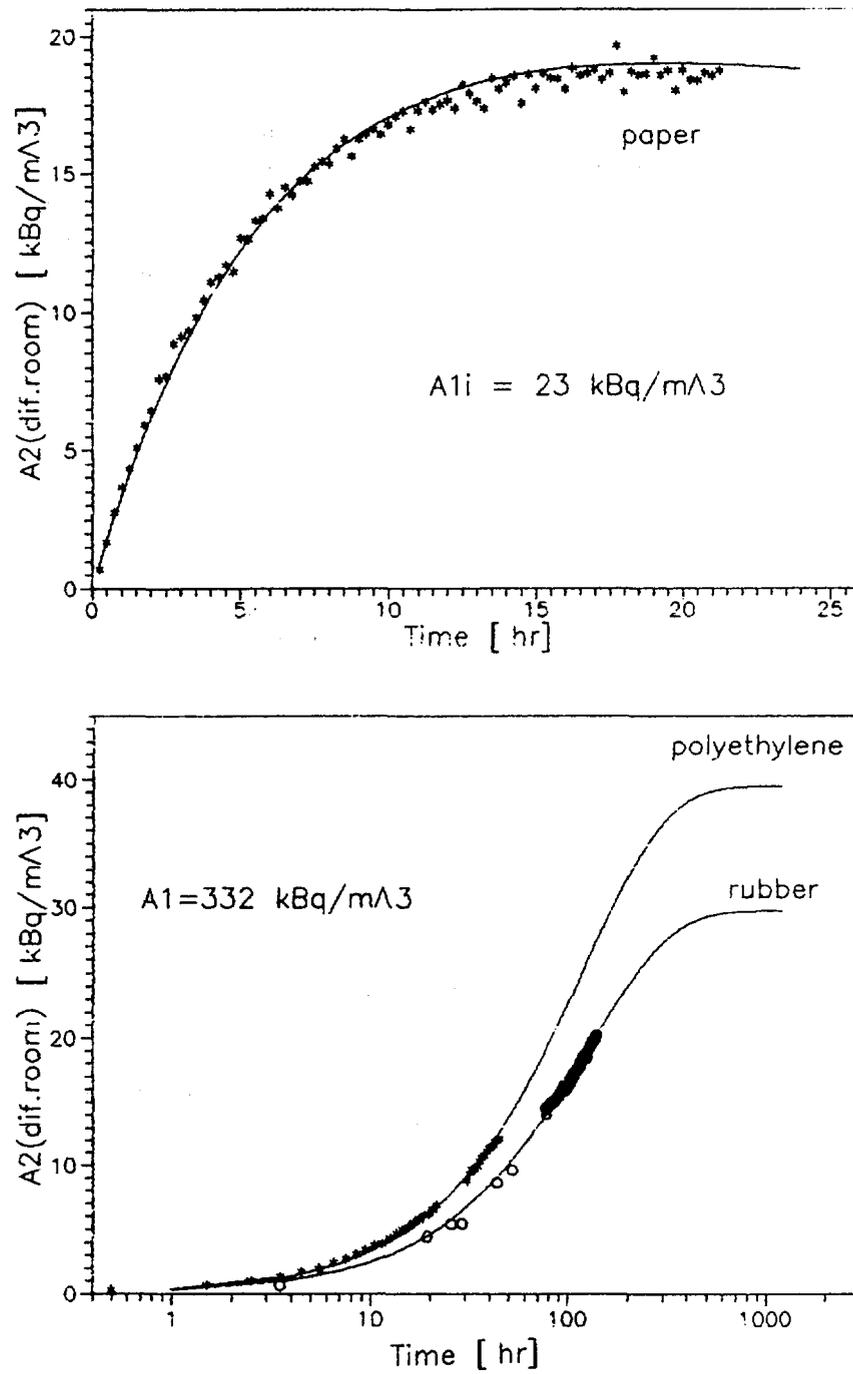
for initial activity  $A_{01}$  in volume  $V_1$ , constant activity  $A_2$  and  $\phi=0$ .

**Experimental results**

Six samples were studied for radon diffusion. The thicknesses of barriers were from 0.012 mm to 2 mm, the diffusion area was 16 cm<sup>2</sup> or 200 cm<sup>2</sup> and the volume  $V_2$  was 30 dm<sup>3</sup>. The following values for  $D$  are in Table 1 and they were obtained using the above given expressions and the data from measurements.

**Tab.1** Calculated diffusion coefficients tested barriers

| Barriers        | D [m <sup>2</sup> /s]        | R [mm]    | h [mm] |
|-----------------|------------------------------|-----------|--------|
| Filter paper    | (1.25±0.13).10 <sup>-7</sup> | 244±14    | 0.17   |
| Rubber          | (4.5±0.3).10 <sup>-10</sup>  | 14.3±0.2  | 1.4    |
| Polyethylene LD | (4.26±0.1).10 <sup>-12</sup> | 1.47±0.05 | 0.012  |
| Polyethylene HD | (2.1±0.3).10 <sup>-12</sup>  | 1.0±0.1   | 0.55   |
| Glasslaminat    | (1.92±0.4).10 <sup>-12</sup> | 0.96±0.1  | 2.0    |
| Polypropylene   | (5.5±0.6).10 <sup>-13</sup>  | 0.5±0.03  | 0.09   |



**Fig. 2** Radon gas activities in diffusion room for different barriers: experimental - poits  
theoretical - lines

Figure 2 shows the growth of radon concentrations measured with radon monitor and calculated from the above written equations in diffusion room for different barriers. The overall relative uncertainty of measured diffusion coefficients is better than 20 %.

### **Conclusion**

The procedures used in our experiments are useful for study of diffusion ability of radon and thoron in barriers and for determination diffusion parameters from short term measurements. We should like to continue in the studies of diffusion parameters especially for performing a comparison of radon and thoron diffusion parameters. In the future we will also take into account the influence of environmental conditions on radon permeability through the barrier and the estimation of radon and thoron concentrations by passive track detectors.

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## THE CONCENTRATION OF NATURAL RADIONUCLIDES IN VARIOUS TYPES OF BUILDING MATERIALS IN SLOVAKIA

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### Introduction

Natural radiation is the main source of exposure to humans. The basic raw materials, generally used in construction industry, contain natural radionuclides, content of which reflects their natural origin and the geological conditions at their site of their production. In the last time, most building materials are manufactured from secondary raw materials with higher concentration of natural radionuclides. The estimation of the  $^{226}\text{Ra}$  content as well as the  $^{232}\text{Th}$  and  $^{40}\text{K}$  concentrations in building materials and products is essential for the evaluation of the external gamma ray contribution to the exposure. The building materials with high values of  $^{226}\text{Ra}$  coupled with the pronounced porosity of the final products, make them potential indoor Rn sources.

According to the radiation protection regulations in Slovakia, our institute regularly investigate the concentrations of natural radionuclides in samples of building raw materials and products. This work is performed in close cooperation with the building material producers, through the Technical and testing institute of building trade in Bratislava.

### Method

The concentration of the natural radionuclides is determined by the gammaspectrometry analysis using 130 cm<sup>3</sup> HPGe detector (30% detection efficiency, 1.95 keV/1.33 MeV resolution) and MCA LIVIUS 2000.

$^{226}\text{Ra}$  and  $^{232}\text{Th}$  are assessed through their progeny photopeaks  $^{214}\text{Bi}$  (609 keV),  $^{214}\text{Pb}$  (295 keV, 351 keV),  $^{228}\text{Ac}$  (338 keV, 911 keV) and  $^{212}\text{Pb}$  (238keV). The specific activity of both nuclides is determined as weighted average of their photopeaks.  $^{40}\text{K}$  is measured directly via its 1460 keV peak. The radium equivalent activity is calculated from specific activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ . All samples are measured in 4 geometry (Marinelli type sample). The building materials and products are milled and screened with 2-3 mm sieve. After drying the samples are stored in 450 cm<sup>3</sup> sealed polyethylene container for 30 days ingrowth period.

The results of analysis are corrected to the background distribution and to the selfabsorption in the volume of the samples. The efficiency calibration is realized using the reference sources distributed by IAEA in Vienna and by the Institute for Radionuclide Production in Prague.

### Results

The measured activity concentrations of the building materials are given in the Table 1. There are shown the minimum and maximum values for different investigated materials.

In cooperation with the Institute of living was analysed 60 samples of building products (panel measured from inner- and outside of the wall) and the values are expressed in Table 2,3. The mean values of specific activities for the outside of the wall are:  $^{40}\text{K}$ : 363.3 Bq.kg<sup>-1</sup>,  $^{226}\text{Ra}$ : 35.9 Bq.kg<sup>-1</sup>,  $^{232}\text{Th}$ : 28.9 Bq.kg<sup>-1</sup>. For the inside panel, those values are only  $^{40}\text{K}$ : 273.0 Bq.kg<sup>-1</sup>,  $^{226}\text{Ra}$ : 15.9 Bq.kg<sup>-1</sup> and  $^{232}\text{Th}$ : 15.7 Bq.kg<sup>-1</sup>. The values of annual mean effective dose rates of gamma radiation from these samples are introduced in Table 4,

**Tab. 1** The specific activities of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and the specific volume activity of  $^{226}\text{Ra}$  in the various types of building materials in Slovakia

| materials  | samp | A [Bq/kg]       |      |                   |      |                   |      |                  |      |
|------------|------|-----------------|------|-------------------|------|-------------------|------|------------------|------|
|            |      | $^{40}\text{K}$ |      | $^{226}\text{Ra}$ |      | $^{232}\text{Th}$ |      | $a_{\text{Ekv}}$ |      |
|            |      | min.            | max. | min.              | max. | min.              | max. | min.             | max. |
| cement     | 22   | 182             | 352  | 16                | 73   | 12                | 27   | 50               | 113  |
| stone      | 42   | 3               | 2441 | 3                 | 65   | 1                 | 112  | 4                | 443  |
| fly-ash    | 33   | 174             | 903  | 67                | 318  | 48                | 159  | 190              | 535  |
| light con. | 18   | 72              | 652  | 11                | 265  | 10                | 285  | 50               | 630  |
| slag       | 6    | 166             | 395  | 41                | 124  | 41                | 106  | 105              | 287  |
| dross      | 3    | 179             | 536  | 60                | 215  | 48                | 56   | 135              | 311  |
| sand       | 9    | 208             | 419  | 8                 | 21   | 8                 | 29   | 31               | 93   |
| calsilox   | 5    | 330             | 416  | 49                | 70   | 34                | 70   | 120              | 191  |
| dolomit    | 9    | 1               | 198  | 5                 | 18   | 1                 | 13   | 6                | 21   |
| alumino-   |      |                 |      |                   |      |                   |      |                  |      |
| phosphate  | 5    | 185             | 869  | 24                | 116  | 37                | 213  | 87               | 398  |
| other      | 28   | 18              | 2488 | 5                 | 108  | 1                 | 118  | 16               | 394  |

**Tab. 2** The specific activities of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and the specific volume activity of  $^{226}\text{Ra}$  in innerside of the wall in Slovakia

| City           | A [Bq/kg] |          |          |                  |
|----------------|-----------|----------|----------|------------------|
|                | a(K)      | a(Ra)    | a(Th)    | $a_{\text{Ekv}}$ |
| Vranov         | 408,6±41  | 37,0±5,6 | 36,0±5,4 | 117,5±16,0       |
| Bratislava     | 400,5±41  | 25,7±3,9 | 23,0±3,4 | 89,1±11,7        |
| Levice         | 275,8±28  | 36,6±5,5 | 27,7±4,2 | 95,2±13,2        |
| Bardejov       | 472,6±47  | 28,3±4,2 | 28,9±4,3 | 105,4±3,7        |
| Rožňava        | 246,0±25  | 30,5±4,6 | 22,7±3,4 | 80,3±11,0        |
| B. Bystrica    | 343,9±35  | 36,4±5,5 | 25,4±3,8 | 98,0±13,3        |
| B. Bysrtica    | 204,1±20  | 15,0±3,0 | 8,1±2,4  | 42,8±7,8         |
| Žiar n. Hronom | 403,8±41  | 53,8±8,1 | 36,5±5,5 | 134,5±18,6       |
| Žiar n. Hronom | 443,0±28  | 20,0±2,0 | 25,0±2,0 | 90,0±6,0         |
| Krompachy      | 596,9±60  | 28,2±4,2 | 39,3±5,9 | 129,1±16,8       |
| Žilina         | 263,4±27  | 61,5±9,3 | 28,1± 2  | 119,6±17,0       |
| Žilina         | 233,0±17  | 56,0±4,0 | 20,0±3,0 | 100,0±6,0        |
| Piešťany       | 345,7±35  | 29,9±4,5 | 34,5±5,2 | 103,2±14,1       |
| Kežmarok       | 371,5± 37 | 17,1±2,7 | 26,4±4,0 | 82,3±11,0        |
| Nitra          | 440,0±45  | 62,0±9,3 | 52,3±7,8 | 165,8±23,0       |

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**Tab. 3** The specific activities of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and the specific volume activity of  $^{226}\text{Ra}$  in outside of the wall in Slovakia

| City        | A [Bq/kg] |          |          |                  |
|-------------|-----------|----------|----------|------------------|
|             | a(K)      | a(Ra)    | a(Th)    | $a_{\text{Ekv}}$ |
| Vranov      | 251.5±26  | 24.8±3.8 | 23.0±3.5 | 75.4±10.5        |
| Bratislava  | 283.8±28  | 15.7±2.4 | 13.0±1.2 | 52.6±6.3         |
| Levice      | 277.2±28  | 13.4±2.0 | 11.6±1.7 | 51.9±6.6         |
| Bardejov    | 273.6±28  | 18.5±2.8 | 17.1±2.6 | 63.6±8.5         |
| Rožňava     | 483.6±48  | 20.0±3.0 | 34.0±5.1 | 104.1±13.6       |
| B. Bystrica | 264.1±26  | 13.1±2.0 | 13.4±2.0 | 52.7±6.8         |
| B. Bysrtica | 150.2±16  | 11.8±2.2 | 8.0±0.3  | 34.8±6.5         |
| Krompachy   | 442.9±45  | 16.2±2.4 | 22.8±2.0 | 83.0±8.8         |
| Žilina      | 366.0±37  | 17.5±2.6 | 17.7±2.7 | 71.3±9.2         |
| Žilina      | 127.8±15  | 29.6±4.4 | 13.1±3.3 | 57.1±9.9         |
| Piešťany    | 240.0±24  | 13.2±2.0 | 12.3±1.8 | 49.4±6.3         |
| Kežmarok    | 245.0±35  | 13.1±2.0 | 19.1±2.9 | 66.8±8.7         |
| Nitra       | 178.1±18  | 7.5± .2  | 8.2±2.1  | 33.1±5.4         |
| Nitra       | 284.5±29  | 13.4±1.8 | 11.7±1.8 | 2.6±6.9          |

**Tab. 4** The values of annual mean effective dose rates of gamma radiation from samples of inner- and outside of the wall in Slovakia

| City           | $\dot{H}$ [mSv.y <sup>-1</sup> ] |           |
|----------------|----------------------------------|-----------|
|                | outside                          | innerside |
| Vranov         | 0,2961                           | 0,1903    |
| Bratislava     | 0,2208                           | 0,1387    |
| Levice         | 0,2408                           | 0,1268    |
| Bardejov       | 0,2617                           | 0,1581    |
| Rožňava        | 0,2024                           | 0,2609    |
| B. Bystrica    | 0,2452                           | 0,1298    |
| B. Bysrtica    | 0,1046                           | 0,0864    |
| Žiar n. Hronom | 0,3390                           | 0,1059    |
| Žiar n. Hronom | 0,2217                           | -         |
| Krompachy      | 0,3215                           | 0,2045    |
| Žilina         | 0,3021                           | 0,1752    |
| Žilina         | 0,2100                           | 0,1441    |
| Piešťany       | 0,2612                           | 0,1217    |
| Kežmarok       | 0,1807                           | 0,1462    |
| Nitra          | 0,4217                           | 0,0813    |
| Nitra          | -                                | 0,1285    |

## RADIOMETRIC DETERMINATION OF $^{226}\text{Ra}$ , $^{228}\text{Ac}$ AND $^{40}\text{K}$ IN FLY ASHES AND BUILDING MATERIALS

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It is well known that coal and oil tend to concentrate radioelements including radium, thorium, uranium and potassium-40. Coal- and oil-burning plants, therefore, produce fly-ash with greater radioelements content. Fly-ash is used by construction industry for building material (panel) production. Exposure rates measured in houses built from fly-ash panels are greater than in the case of other building materials<sup>1,2</sup>. This was the reason why we studied the radioactivity of Slovak fly-ashes and building materials produced from fly-ashes.

### Experimental

For measuring fly-ash and building material samples Marinelli-type sample containers were used, made of 0.3 mm thick electrolytic copper. The dimensions of sample containers were as follows: external diameter 14 mm, internal diameter 8 mm, height 105 mm, depth 75 mm. Before container filling, all of the samples were homogenized and their density was established. After the containers were filled, they were closed airtight for 20 d to reach radioactive equilibrium.

A semiconductor Ge(Li) detector with 20% detection efficiency and with resolution of 2 eV (at 1.33 MeV) was used for measurement. The detector was placed in a 100 mm thick lead shielder. The analogue digital part of the spectrometer consisted of a pulse height analyzer, Canberra type CI 8180, a preamplifier type CI 2001 A and of an amplifier type CI 2010. The identification of particular components was carried out by means of a PDP 1014 computer. The long term stability was controlled and the background of the detection assembly was subtracted. The calibration of the measuring system was realized by means of a set of reference source (ÚVVR, Prague).

The measuring time of every sample was 3000 s. By calculation of the specific activities, corrections to absorption were realized<sup>3</sup>.

### Results and Discussion

The results obtained are shown in Table I.

From Table I it is clear that the different origin of coals combusted results in significant differences in specific activities of  $^{226}\text{Ra}$  and  $^{228}\text{Ac}$  of the measured fly-ashes. The knowledge of the specific activity of selected nuclides contained in fly-ashes is, therefore, very important and in specific cases can indicate the possibilities of their further technological use.

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**Tab. 1** Specific activities of  $^{226}\text{Ra}$ ,  $^{228}\text{Ac}$  and  $^{40}\text{K}$  of fly-ash and building material (panel) samples

| Sample  | $^{226}\text{Ra}$ |              | $^{228}\text{Ac}$ |              | $^{40}\text{K}$ |              |
|---|-------------------|--------------|-------------------|--------------|-----------------|--------------|
|   | A [Bq/kg]         | $\sigma$ [%] | A [Bq/kg]         | $\sigma$ [%] | A [Bq/kg]       | $\sigma$ [%] |
| 70 Roberton<br>Šaštín Srtáže<br>(fly-ash)             | 9                 | 50           | 18                | 30           | 280             | 10           |
| Börský Mikuláš<br>(fly-ash)                           | 17                | 30           | 15                | 50           | 615             | 10           |
| Vranov n.Topľou<br>(fly-ash)                          | 79                | 10           | 64                | 15           | 589             | 10           |
| ENO-2 Nováky<br>(fly-ash)                             | 85                | 20           | 75                | 20           | 720             | 10           |
| Niklova huť Sereď<br>(fly-ash)                        | 107               | 10           | 87                | 15           | 510             | 10           |
| Porobetón Bratislava<br>(fly-ash)                     | 95                | 15           | 45                | 25           | 530             | 10           |
| Porobetón Sereď<br>(fly-ash)                          | 90                | 15           | 46                | 20           | 563             | 10           |
| Šaštín Stráže<br>70 Roberton<br>(building mat.-panel) | 18                | 30           | -                 | -            | 360             | 10           |
| Zem. Kostofany III.<br>(building mat.-panel)          | 62                | 20           | 53                | 20           | 520             | 10           |
| Porobetón, Sereď<br>(building mat.- panel)            | 51                | 20           | -                 | -            | 263             | 20           |
| Porobetón, Zem.<br>Kostofany (building<br>mat.-panel) | 54                | 25           | 25                | 30           | 662             | 10           |



## CYTOGENETIC EFFECTS OF TRITIUM INCORPORATED INTO DNA OF HUMAN LYMPHOCYTES

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### **Introduction**

Tritium produced by nuclear industry is deposited in the environment and a small proportion of it is eventually incorporated into the genetic material of cells. At high risk to the genetic material are the tritiated precursors of DNA used mainly in laboratory experimental work (Faerber et al. 1990).

There is available information about the relation of dose to the response using the concentration in the incubation medium for expressing the dosage of tritium and using the number of chromosomal aberrations (CA) as the response endpoint after high doses of tritium incorporated into DNA in cells in vitro (Bond et al. 1966, Pelliccia et al. 1988, Faerber et al. 1990). The physical doses expressed in units of Gy, however, may widely vary in such experiments as the specific radioactivity of the labelled thymidine molecules from experiment to experiment was very variable. Moreover, the information about physical dose-response relationship for low dose tritium incorporated into DNA of human lymphocytes, in following CA as endpoints of response are scarce.

In the reported in vitro experiments the numbers of chromosomal aberrations (CA) in correlation to the physical dose as assessed by determining the specific radioactivity of DNA have been followed in in vitro human lymphocytes from adult donors.

### **Methods**

Lymphocytes from healthy adult donors of age from 20 to 59 years, of both sexes (24 males and 20 females) were isolated from blood by centrifugation. After washing the cells were irradiated from tritium incorporated during in vitro incubation in phytohemagglutinin (Wellcome) containing medium with tritium labelled thymidine ( $^3\text{HTdR}$ , specific activity 1950 GBq/mM, UVVVR Praha). The concentration of tritium in medium was changed in five intraindividual samples of lymphocytes from 65 to 948 Bq/ml.

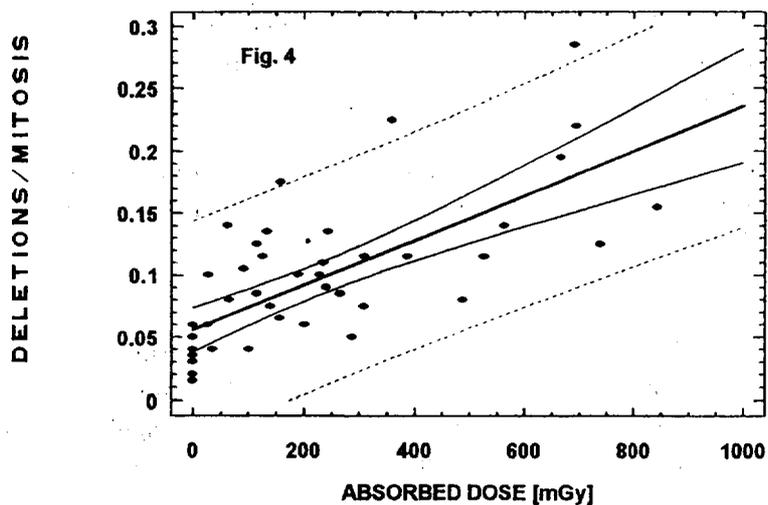
Slides for standard CA counting have been done from every sample 48 hours after the begin of the cultivation. The CA were counted in at least 200 metaphases on each slide. Parallel samples of lymphocytes served for preparing smears for autoradiography to determine the labeling index. Other parallel samples were used for the determination of tritium concentration in DNA by the diphenylamine method, as well as for determination of the specific radioactivity in lymphocyte DNA by scintillation counting. The dose absorbed in DNA was estimated using the conversion factor implicating that 37 kBq of tritium uniformly distributed per gram of tissue of unit density delivers a dose rate of 12.14 mrad/hour (Cronkite et al. 1973).

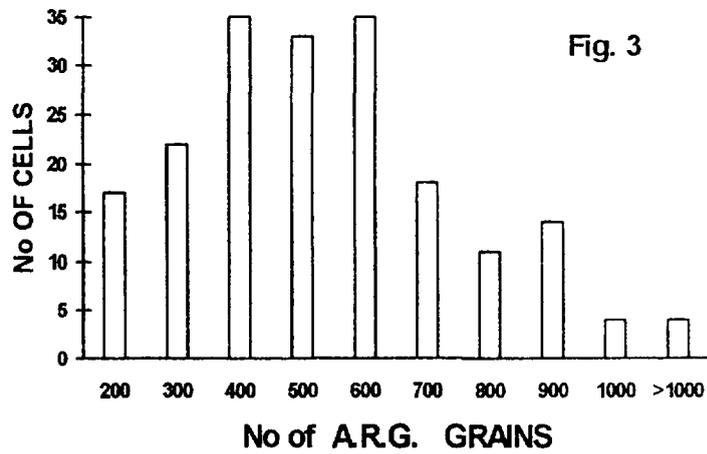
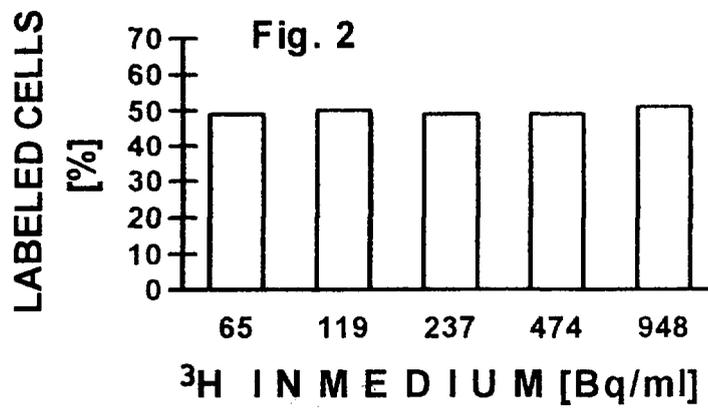
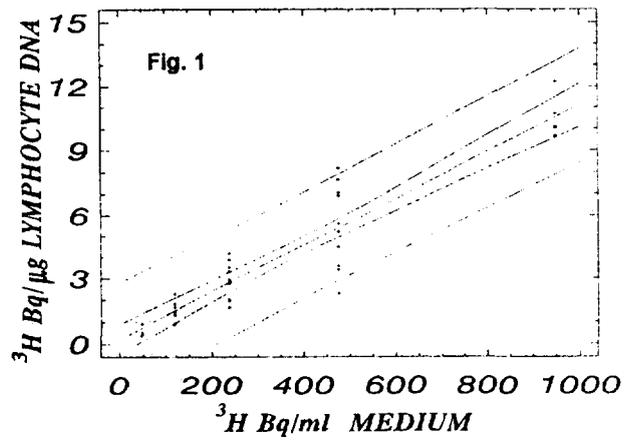
### **Results**

The dependence of the concentration of tritium in the lymphocyte DNA upon its concentration in medium is on Fig.1. A linear relationship between these two quantities in the used range of concentrations could be found. It follows from the graph that, at the specific tritiated thymidine radioactivity of 1950 GBq/mM, the rise of the specific radioactivity of DNA by about 1 Bq/mg required to raise the concentration of the  $^3\text{HTdR}$  in medium by approximately 100 Bq/ml.

The autoradiographic analysis showed that only about one half of the cells were labelled as is shown on Fig.2 where labelling indexes of the lymphocyte population after incubation in various concentrations of  $^3\text{HTdR}$  in medium were plotted. The labelling indexes of populations of lymphocytes incubated in did not show significant variation. An example of the frequency distribution of the tagged cells according to the labelling intensity is shown on Fig.3. This distribution may be better approximated by a log-normal curve, than by a Gaussian. It shows an uneven intensity of labelling in the labelled proportion of cells.

A plot of the number of CA of the deletion type (chromatid deletions, chromosome and chromatid fragments, chromosome and chromatid breaks) against the estimated absorbed dose is shown on Fig 4. From this plot it follows that at the conditions used the dose of 100 mGy absorbed from tritium causes about 0,1 deletions per cell. For doubling the number of CA to about 0,2 deletions per cell, five times higher absorbed dose (i.e. about 500 mGy) is necessary.





### Discussion

The results of this study agree quite well with that published in the review of Bond et al.(1966). In this review the dose response curve for CA in Chinese hamster cells after tritium irradiation by doses up to 5.5 Gy has been published.

As already mentioned, the doubling of the response requires to raise the dose five times. This causes that the slope of the dose-response curve is much flatter than could be expected from physical point of view and from experiments with single dose acute irradiation. The flatter dose-response curve reflects, first, that damaged cells die and undergo cell lysis so that they would not be detectable in the prepared slides as cells with CA. Second, it may reflect also the quick repair processes in many cells replicating DNA during the prolonged incubation period, during which these cells may diminish the number of DNA chain breaks so that microscopically visible aberrations do not appear. Both these features are based upon the natural processes governing thymidine metabolism in cells, its temporal character that may be typical for all processes of internal contamination. In comparing with the clastogenic effects of acute external irradiation these processes cause that the dose absorbed by the cell population rises slowly, thus expressing a subchronic character of response.

### Conclusions

- 1) The contamination of cells by precursors of nucleic acids - like tritiated thymidine - causes an uneven distribution of doses in the cell population. A proportion of the population of cells remains unlabelled.
- 2) the dose-response curve is flat showing signs of loss of heavily damaged cells and signs of repair of damage. Both these signs are based on the nature of biological processes which lead to internal contamination of cells and to expression of effects in terms of numbers of chromosomal aberrations.

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## THE RADIOPHARMACEUTICALS IN NUCLEAR MEDICINE DIAGNOSTIC PROCEDURES AND THE PROBLEM OF RADIATION PROTECTION

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### Introduction

Many radionuclides used in nuclear medicine ( $^{67}\text{Ga}$ ,  $^{99\text{m}}\text{Tc}$ ,  $^{111}\text{In}$ ,  $^{123}\text{I}$ ,  $^{125}\text{I}$ ,  $^{201}\text{Tl}$ ) decay by electron capture (EC) and/or internal conversion (IC) and the result is the emission of a large number of low energy electrons having ranges of subcellular dimension. Radionuclides that emit Auger electrons can be extremely radiotoxic depending on the subcellular distribution of the radiochemical (1,4,11). Calculation of the absorbed dose and equivalent dose to tissues from incorporated Auger emitters is important for risk assessment.

The ICRP 26 (7) in the earlier recommendations defined the dose equivalent H according to the relationship  $H = D \cdot Q \cdot N$ , where D is absorbed dose, Q is quality factor and N is product of all other modifying factors (e.g., dose rate). They also recommended  $N=1$  for all situations and  $Q=1$  for all electrons. Accordingly, application of these recommendations to the Auger electron emitters would lead to dose equivalent that is simply equal to the absorbed dose. The extreme radiotoxicity observed for a variety of Auger emitters is not accounted for by this approach. There is now a wealth of informations from *in vitro* and *in vivo* studies concerning the radiotoxicity of Auger electron emitters (3,4,9,10). All of these experimental data show a striking dependence of the cytotoxicity on the subcellular distribution as might be expected given the highly localized energy deposition around the decay site. More specifically, when Auger emitters are situated outside the cell, or in the cytoplasm of the cell, only effects similar to radiation of low linear energy transfer (LET) are observed with values of relative biological effectiveness (RBE) about 1. On the other site, localization of these radionuclides within the DNA or in close neighbourhood of DNA in the cell nucleus can produce extreme radiotoxicity (RBE about 7-9 for cell killing).

The ICRP 60 (8) updated their recommendations and newly defined the tissue equivalent dose  $H_T = \sum_R W_R \cdot D_{T,R}$ , where  $W_R$  is the radiation weighting factor and  $D_{T,R}$  is the absorbed dose to tissue T, from R-th component of radiation.

No guidance is provided regarding radiation weighting factors for Auger emitters. In alluding to this problem, it was only indicated that microdosimetry techniques are required to establish  $W_R$  for Auger emitters.

It should be noted, however, that until radiosensitive targets within the cell nucleus are well defined and both the microscopic distribution of the radionuclides relative to the targets and time dependence of distribution are known, it is unlikely that meaningful microdosimetry calculations can be performed, that predict the extremely varied biological effects caused by Auger emitters.

So the ICRP 60 recommends that  $W_R$  values be based primarily on experimental RBE values of radiation.

The recent report (6) by American Association of Physicists in Medicine (AAPM) recommends a radiation weighting factor  $W_R$  of 20 for stochastic effect caused by Auger electrons.

Howell et al. (5) propose that equivalent dose specifically for Auger electrons may be expressed as:

$$H_{T,R(\text{Auger})} = [1 + f_0(W_{R(\text{Auger})} - 1)] \sum_{R(\text{Auger})} D_{T,R(\text{Auger})} \quad (1)$$

where  $f_0$  is the fraction of radioactivity in organ bound to DNA,  $D_{T,R(Auger)}$  is the absorbed dose in tissue T from Auger electrons.

In view of these recommendations, it is important to reevaluate equivalent dose from Auger electrons emitters. The expression (1) allows calculation of the equivalent dose for Auger electrons emitters given the mean tissue dose from the different types of radiation and the subcellular distribution of the radionuclide in the tissue.

### Methods

#### 1. Absorbed dose per unit cumulated activity.

The mean self-absorbed dose per unit cumulated activity  $S_{T \leftarrow T}$  to the tissue T is given as:

$$S_{T \leftarrow T} = \sum_{R(Auger)} S_{T \leftarrow T, R(Auger)} + \sum_{R(Other)} S_{T \leftarrow T, R(Other)} = \frac{1}{m_T} \left\{ \sum_{R(Auger)} \Gamma_{R(Auger)} \cdot \Phi_{T \leftarrow T, R(Auger)} + \sum_{R(Other)} \Gamma_{R(Other)} \cdot \Phi_{T \leftarrow T, R(Other)} \right\} \quad (2)$$

where  $\sum_{R(Auger)} S_{T \leftarrow T, R(Auger)}$  and  $\sum_{R(Other)} S_{T \leftarrow T, R(Other)}$  are mean self-absorbed dose per unit cumulated activity in tissue T from Auger electrons and others radiation,  $m_T$  is mass of tissue T and  $\Gamma_{R(Auger)}$ ,  $\Phi_{T \leftarrow T, R(Auger)}$  are mean energy emitted per nuclear transition and self-absorbed fraction for the R-th Auger electron component, respectively.  $\Gamma_{R(Other)}$ ,  $\Phi_{T \leftarrow T, R(Other)}$  are the same for the R-th radiation component other than Auger electrons.

#### 2. Equivalent dose per unit cumulated activity.

Although the absorbed dose is useful dosimetric quality in radiological protection, the biological effects of radiation also are known to depend on the radiation type and energy of radiation. Therefore, in order to predict biological effect, there is definition of equivalent dose as the weighted absorbed dose.

The equivalent dose to the tissue T per unit cumulated activity is:

$$S_{T \leftarrow T}^H = [1 + f_0(W_{R(Auger)} - 1)] \sum_{R(Auger)} S_{T \leftarrow T, R(Auger)} + \sum_{R(Other)} W_{R(Other)} S_{T \leftarrow T, R(Other)} \quad (3)$$

The AAPM (as mentioned earlier) recommended the value of  $W_{R(Auger)}$  for stochastic effect to be 20.

#### 3. Absorbed dose and equivalent dose.

The mean self-absorbed dose to tissue T from activity in tissue T is simply:

$$D_{T \leftarrow T} = A_T S_{T \leftarrow T} \quad (4)$$

The quantity of  $A_T$  is the cumulated activity in tissue T. In terms of the equivalent dose per unit cumulated activity, the equivalent dose to the tissue T from activity in this tissue is simply:

$$H_{T \leftarrow T} = A_T S_{T \leftarrow T}^H \quad (5)$$

### Results and discussion

To demonstrate the effect of the radiation weighting factor for Auger electrons on equivalent dose according to the work of Goddu et al. (2), the Table 1 gives the equivalent dose to tissue T per unit cumulated activity  $S_{T \leftarrow T}^H$  for various radionuclides commonly used in nuclear medicine ( $^{67}\text{Ga}$ ,  $^{123}\text{I}$ ,  $^{125}\text{I}$ ,  $^{99m}\text{Tc}$ ,  $^{111}\text{In}$ ,  $^{201}\text{Tl}$ ). The quantities  $S_{\text{Testes} \leftarrow \text{Testes}}^H$  and  $S_{\text{Liver} \leftarrow \text{Liver}}^H$  are tabulated and compared with  $S_{\text{Testes} \leftarrow \text{Testes}}$  and  $S_{\text{Liver} \leftarrow \text{Liver}}$ . Two values of  $f_0$  are considered 0.5 and 1.0 and the ratio of  $S_{\text{Testes} \leftarrow \text{Testes}}^H / S_{\text{Testes} \leftarrow \text{Testes}}$  and  $S_{\text{Liver} \leftarrow \text{Liver}}^H / S_{\text{Liver} \leftarrow \text{Liver}}$  are presented. These ratios are related to

the expected RBE for the stated values of  $f_0$ . The data on the Table 1 show that the equivalent dose is highly dependent on the fraction of the activity bound to the DNA, illustrating the importance of subcellular distribution of Auger electron emitters in assessing the equivalent dose.

### Conclusions

The presented approach represents a practical step toward the estimation of equivalent dose for incorporated Auger electron emitters, an aspects that has not been given adequate consideration so far. Given the widespread use of this class of radionuclides in nuclear medicine and in biomedical research, the formalism and practical calculation presented here may be of value to assessing the risk associated with exposure to this radionuclides (in diagnostic nuclear medicine procedures), as well as predicting their therapeutic efficiency.

**Tab. 1A, 1B** Ratio of equivalent dose and mean absorbed dose per unit cumulated activity distributed in human testes and liver from Auger electrons emitters.

1A

| Radionuclide      | Human Testes         |                        |                              |                        |                              |
|-------------------|----------------------|------------------------|------------------------------|------------------------|------------------------------|
|                   | $S_{T<T}$<br>Gy/Bq s | 50% in DNA             |                              | 100% in DNA            |                              |
|                   |                      | $S^H_{T<T}$<br>Sv/Bq s | $S^H_{T<T}/S_{T<T}$<br>Sv/Gy | $S^H_{T<T}$<br>Sv/Bq s | $S^H_{T<T}/S_{T<T}$<br>Sv/Gy |
| <sup>67</sup> Ga  | 2.10E-13             | 4.89E-13               | 2.3                          | 7.68E-13               | 3.7                          |
| <sup>123</sup> I  | 1.93E-13             | 5.19E-13               | 2.7                          | 8.44E-13               | 4.4                          |
| <sup>125</sup> I  | 1.44E-13             | 6.78E-13               | 4.7                          | 1.21E-12               | 8.4                          |
| <sup>111</sup> In | 2.79E-13             | 5.83E-13               | 2.1                          | 8.88E-13               | 3.2                          |
| <sup>99m</sup> Tc | 1.03E-13             | 1.47E-13               | 1.4                          | 1.92E-13               | 1.9                          |
| <sup>201</sup> Tl | 2.32E-13             | 8.37E-13               | 3.6                          | 1.44E-12               | 6.2                          |

1B

| Radionuclide      | Human Testes         |                        |                              |                        |                              |
|-------------------|----------------------|------------------------|------------------------------|------------------------|------------------------------|
|                   | $S_{T<T}$<br>Gy/Bq s | 50% in DNA             |                              | 100% in DNA            |                              |
|                   |                      | $S^H_{T<T}$<br>Sv/Bq s | $S^H_{T<T}/S_{T<T}$<br>Sv/Gy | $S^H_{T<T}$<br>Sv/Bq s | $S^H_{T<T}/S_{T<T}$<br>Sv/Gy |
| <sup>67</sup> Ga  | 6.01E-15             | 1.14E-14               | 1.9                          | 1.69E-14               | 2.8                          |
| <sup>123</sup> I  | 6.41E-15             | 1.27E-14               | 2.0                          | 1.91E-14               | 3.0                          |
| <sup>125</sup> I  | 4.09E-15             | 1.45E-14               | 3.5                          | 2.49E-14               | 6.1                          |
| <sup>111</sup> In | 1.08E-14             | 1.68E-14               | 1.6                          | 2.27E-14               | 2.1                          |
| <sup>99m</sup> Tc | 3.69E-15             | 4.55E-15               | 1.2                          | 5.41E-15               | 1.5                          |
| <sup>201</sup> Tl | 5.96E-15             | 1.77E-14               | 3.0                          | 2.95E-14               | 5.0                          |

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## TO THE RADIOTOXICITY OF $^{99m}\text{Tc}$ RADIOPHARMACEUTICALS

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Technetium-99m is considered to be a radionuclide of choice for imaging in nuclear medicine because of its short physical half life ( $T_p=6\text{h}$ ), isometric transition (absence of energetic beta radiation), 140.5 - keV gamma rays, which are efficiently detected by a NaI gamma camera, and ready availability at high specific activities using a generator.

So in a view of widespread use of  $^{99m}\text{Tc}$  in nuclear medicine as mentioned above, and because the dependence of radiotoxicity of Auger emitters on their subcellular distribution, it is important to ascertain the RBE values for  $^{99m}\text{Tc}$  radiopharmaceuticals.

It has been shown (1) that the experimental RBE value is simply the dose - weighted sum of the RBE values of the different groups of radiations emitted by the radionuclide. However, the RBE of the Auger electrons must also be weighted by the fraction of organ activity that is bound to DNA (3). Accordingly, an expression for the expected RBE for  $^{99m}\text{Tc}$  compounds can be written as follows:

$$\text{RBE}_{\text{expected}}(^{99m}\text{Tc}) = f_{\text{photons}} \text{RBE}_{\text{photons}} + f_{\text{CE}} \text{RBE}_{\text{CE}} + f_{\text{Auger}} f_{\text{DNA}} \text{RBE}_{\text{Auger}} \quad (1)$$

where  $f_{\text{photons}}$ ,  $f_{\text{CE}}$ ,  $f_{\text{Auger}}$  are the fraction of the tissue absorbed dose resulting from photons, conversion electrons (CE) and Auger electrons respectively, and  $f_{\text{DNA}}$  is the fraction of the tissue cumulated activity bound to DNA.

It is generally recognized that  $\text{RBE}_{\text{photons}}$  and  $\text{RBE}_{\text{CE}}$  is unity. If we assume that the Auger electrons emitted in the decay of  $^{99m}\text{Tc}$  are as effective as the  $^{125}\text{I}$  Auger electrons in causing the biological damage as recommended by AAPM Task Group on Auger Electron Dosimetry (3), then  $\text{RBE}_{\text{Auger}}=10$ .

In the case of using spermatogenesis in mouse testes as the experimental model, the values of fractions of tissue absorbed dose are  $f_{\text{photons}} = 0.889$ ,  $f_{\text{CE}} = 0.058$  and  $f_{\text{Auger}} = 0.053$  (4). Therefore, the 4 Auger electrons emitted by  $^{99m}\text{Tc}$  deposit only a small fraction of the testicular absorbed dose and consequently the radiotoxicity (and also RBE) of different  $^{99m}\text{Tc}$  radiopharmaceuticals only very slightly depends on the value of  $f_{\text{DNA}}$  - on fraction of organ activity bound to DNA. This finding is in contradiction with the observed and calculated radiotoxicity of such Auger electrons emitters as  $^{125}\text{I}$ ,  $^{123}\text{I}$ ,  $^{111}\text{I}$  and  $^{201}\text{Tl}$  for which the average number of low energy electrons per decay is 10-15, 11, 8 and 20 - 37 respectively (2), for which the radiotoxicity (and also the RBE) depends on the extent of incorporation into DNA; the larger fraction of organ activity is bound to DNA, the higher is the radiotoxicity of these radionuclides.

When we take the lower ( $f_{\text{DNA}}=0$ ) and the higher ( $f_{\text{DNA}}=1$ ) limit of the DNA bound fraction, the values of RBE of  $^{99m}\text{Tc}$  in mouse testes range from 0.95 to 1.5. This suggests that Auger electrons emitted in the decay of  $^{99m}\text{Tc}$  are not capable of causing extreme toxicity *in vivo*. In closing, if one is interested in RBE at the very low doses encountered in diagnostic nuclear medicine (for diagnostic administrations of  $^{99m}\text{Tc}$ , the sensitive organ received an absorbed dose of order 1 cGy), the RBE values may be substantially higher than what are observed at  $D_{37}$ . Furthermore, RBE values are well known to depend on the biological system and the endpoint measured. For risk assessment it is usual to consider aberration or transformation data where RBE data are typically substantially higher than they are for survival endpoints. For this reason the AAPM Task Group on Auger Electron Dosimetry has recommended (3) a value of 20 for the radiation weighting factor for Auger electrons.

In the conclusion the results of work (4) provide further support for  $^{99m}\text{Tc}$  as the radionuclide for imaging in nuclear medicine.

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## BIOKINETICS OF $^{131}\text{I}$ IN HUMAN ORGANISM

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### Summary

Time evolution of the cumulated activity in human body is one of the key characteristics determining medical impacts of ionizing radiation. In nuclear medicine, so called effective half-life is mostly used for the evolution description. This quantity is usually estimated by fitting a straight line in semi-logarithmic coordinates. Its novel Bayesian estimate was also proposed and its advantageous properties were verified. During extensive tests, it was found that the effective half-life has limited use as the underlying deterministic relationship time - activity can hardly be taken as (mono)exponential. It stimulated the search for a better and still simple model. A quadratic dependence of  $\ln(\text{activity})$  on  $\ln(\text{time})$  was found as an adequate candidate. Preliminary experiments on a restricted set of real data were promising enough to justify its further elaboration.

The paper reports on the progress made in verifying and exploiting this non-standard model. Its potential exploitation in dosimetric tasks is outlined. The core of the paper is positive verification results on an extensive set of real data.

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### 1. Introduction

The amount of the activity administered for diagnostics/therapy in nuclear medicine has to be large enough to reach the administration aims. At the same time, it should be as low as possible in order to minimize a damage of healthy tissues. It calls for a reliable estimation of the energy imparted by ionizing radiation, ie. of the absorbed dose, to the human body (1-10). The well established MIRD (Medical Internal Radiation Dose) methodology (11,12) provides the necessary physical and experimental background. Knowledge of the cumulated activity in the inspected tissues is its main user's input. The cumulated activity is the integral under the time trajectory of the instantaneous activity. In practice, this trajectory has to be reconstructed from a few measurements. It means that a sort of interpolation is needed: a curve from a pre-specified class has to be fitted to noisy measurements.

The quality of the resulting estimate of the cumulated activity is influenced by:

- the class of the fitted curves which results from the deterministic modelling,
- the estimation (fitting) procedure which is dictated by the model of the noise, by the probabilistic modelling.

This paper is devoted to the deterministic modelling of the activity evolution for diagnostic/therapeutic applications of  $^{131}\text{I}$ . A recently proposed quadratic relationship of  $\ln(\text{instantaneous activity})$  to  $\ln(\text{time})$  (13) is evaluated on an extensive set of real data and refined a bit. It outperforms the standard model  $\ln(\text{instantaneous activity})$  taken as an affine function of time characterized by the effective half-life.

Estimation procedures for an adequate probabilistic model are still under the preparation (14).

The paper is organized as follows. After recalling the essence of the MIRD method, the addressed practical problem is formulated. Then, the tested model of the activity kinetics is outlined. Experimental verifications of the model suitability presented afterwards contain the key message of the paper. The concluding remarks list mostly the running research tasks.

## 2. MIRD Method

In nuclear medicine applications, the absorbed dose quantifies the amount of the energy transferred to an inspected (target) tissue from internal sources (source organs) of ionizing radiation. This complex transfer process depends on:

- the type of emitted radiations,
- the amount of energy emitted by the source organs, expressed usually as the product of the number of transmutations per time unit and the (mean) energy emitted per transmutation,
- the mutual target-source organs geometry,
- the type (mass, size) of organs.

MIRD method can be interpreted as an application of the law of energy conservation. The method takes of the burden from practitioners by tabulating adequate conversion factors (so called S-factors) from user's data to doses. Factors are available for a wide set of radionuclides and for various combinations of source and target organs. With this support, the following pieces of knowledge are needed for a practical evaluation of the absorbed doses: 1) radionuclides present in the human body, 2) masses of the inspected target organs, 3) cumulated activities in the source organs.

This paper addresses the estimation related to the third item. For an estimation of the mass, see (15).

## 3. Addressed Problem

When treating thyroid diseases by  $^{131}\text{I}$ , a diagnostic amount of the radionuclide is administered with the aim to estimate abilities of various lesions to accumulate the (radioactive) iodine. The same accumulation ability is supposed during the therapeutic phase. Then, the doses absorbed in the inspected tissues during the therapeutic phase can be estimated and the adequate activity administration determined.

Knowledge of instantaneous and cumulated activities in the therapeutic phase is also required in order to:

- control the radiohygienic regime of patients,
- check the accumulation similarity in both phases.

The cumulated activity  $A_{\text{cum}}$  is a time integral of the instantaneous activities  $A(t)$  for time  $t > \text{application time} = 0$ . Because of medical/technical reasons, the induced counts  $a(t)$  corresponding to activity values  $A(t)$  are measured in several time instants  $t_i$  only (maximum 2-18). In order to provide the needed reliable estimate of the cumulated activity  $A_{\text{cum}}$ , these data have to be used for estimating the whole curve  $A(t)$ ,  $t > 0$ , which is characteristic for the treated patient.

The registered counts  $a(t)$  are related to the activity by

$$a(t) = c \cdot A(t) \cdot \text{noise} \quad [1]$$

where  $c$  denotes the calibration factor which can safely be estimated using an appropriate calibration procedure (15). Under the given measurement conditions, the desired characterization of the whole curve can be obtained only when

- the trajectory  $A(t)$ ,  $t > 0$ , is parameterized by a smooth function containing a few time invariant parameters only;
- the adequate probabilistic noise description is selected.

For algorithmic reasons, the probabilistic modelling is simplified here and logarithmic-normal model is adopted, ie.

$$\ln(a(t)) = \ln(cA(t)) + \text{white normal noise} \quad [2]$$

We concentrate on verification whether the novel model

$$f(t) = \ln(cA(t)) \quad [3]$$

proposed in (13) is suitable in practice.

#### 4. Tested Model

Usually, the affine model is used:

$$f(t) = \ln(cA(t_1)) - \ln(2)(t-t_1)/T_{ef} \text{ for } t > t_1 \quad [4]$$

In this model,  $t_1 > 0$  is the time instant after which fast complex transients of the activity distribution can be neglected.  $T_{ef}$  is the effective half-life parameterizing this model together with  $t_1$  and  $cA(t_1)$ .

Parameters of this model can be simply estimated by least squares. The activity  $A_{cum,1}$  cumulated after  $t_1$  is

$$A_{cum,1} = cA(t_1) \cdot T_{ef} / \ln(2) \quad [5]$$

This model was, however, found to be too crude in a significant portion of the inspected cases (13,16).

Generic activity trajectories resulting from the compartmental models suggest  $f(t)$  as a sum of several exponentials. Such a form is, however, unfeasible with a few measurements available. Thus, a slight deviation from the affine form is only acceptable when searching for a better model.

The shapes of activity trajectories for the reference man (17,18) indicate the following form as a potential candidate:

$$f(t) = k_1 + k_2 \cdot \ln(t) + k_3 \cdot \ln^2(t) - \ln(2) \cdot t / T_p \quad [6]$$

where  $T_p$  is the known physical half-life of the  $^{131}\text{I}$ .

This model has also triple of unknown parameters ( $k_1, k_2, k_3$ ) which can be estimated by least squares. The sensitive choice of  $t_1$  is avoided and the model can be used immediately after the administration. Consequently, no data are lost and no part of the cumulated activity ( $t < t_1$ ) is neglected.

The trajectory  $f(t) = cA(t)$  modelled in this way has the following desirable qualitative properties:

- starts at zero if  $k_2 > 0$  (no activity is cumulated at administration time yet),
- falls to zero faster than the physical decay if  $k_3 < 0$ ,
- possess a single extreme for  $t > 0$  if  $k_3 < 0$ ,
- models explicitly the physical decay so that the individual biological elimination is to be estimated only.

On the other hand, the cumulated activity  $A_{cum,2}$  cannot be found analytically. Nevertheless, the approximate dependence  $A_{cum,2} = A_{cum,2}(k_1, k_2, k_3)$  seems to be easy to construct.

Performed experiments (13) verifying suitability of the model for real data were promising but clearly insufficient. This paper removes this drawback related to the studied model. Experiments has led to a further refinement of this model.

## 5. Experiments

### 5.1 Material available

The raw material used consists of records related to 2000 patients treated for thyroid diseases at the Clinic of Nuclear Medicine, Faculty Hospital Motol, 2<sup>nd</sup> Medical Faculty, Charles University. The data have been gained as a by-product of routine evaluations by a software system JOD (15).

Among the available data, the records containing at least four (greater than the dimension of the estimated parameter) were processed. The selected records contain at most 20 measurements and 1 up to 4 lesions.

### 5.2 Compared models

The studied model M1 (cf. [6])

$$M1: f(t)=\ln(cA(t))=k_1+k_2 \cdot \ln(t)+k_3 \cdot \ln^2(t)-\ln(2) \cdot t/T_p, t>0,$$

is compared with the standard affine model M2 (cf. [4]):

$$M2: f(t)=\ln(cA(t))=\ln(cA(t_1))-\ln(2)(t-t_1)/T_{ef} \text{ for } t>t_1.$$

During experiments, a lot of alternative non-linear functions of time were tested. Among them, the most promising results were gained when  $\ln^2(t)$  in M1 was replaced by a faster-changing functions of time with

$$M3: f(t)=\ln(cA(t))=k_1+k_2 \cdot \ln(t)+k_3 \cdot t^{1/2} \ln(t)-\ln(2) \cdot t/T_p, t>0$$

$$M4: f(t)=\ln(cA(t))=k_1+k_2 \cdot \ln(t)+k_3 \cdot t^{2/3} \ln(t)-\ln(2) \cdot t/T_p, t>0.$$

Results with  $\ln^2$  replaced by  $\ln^3$ ,  $\ln^4$ ,  $t^{1/2}$ , etc. were unsatisfactory.

### 5.3 Exception handling

All models involved have their "natural" range of parameters ( $k_3 < 0$ ,  $0 < T_{ef} < T_p$ , etc.) which should be and could be respected in estimation. In the intermediate research step reported (noise properties are incompletely respected), this prior knowledge was not built in. Nevertheless, the overall performance picture remains unchanged if the cases violating such restrictions are skipped.

Similar robustness is observable if the cases with severe outliers (several hundreds of percent) are omitted.

### 5.4 Judgement of model performance

The models are compared according to their predictive capabilities because of the following reasons:

- they as predictors for radio-hygienic purposes,
- those which are good in the hard extrapolation problem are believed to grasp substantial invariant features of the reality and are expected to suit for estimation of the patient's "invariant", his ability to accumulate activity.

Specifically, let us consider any of the tested sets of trajectories  $f(t,P)$  parameterized by a multivariate parameter  $P$ .

Let  $P_i$  be its estimate gained from data recorded up to and including time  $t_i$ . The prediction  $p_{i+1}$  of the value  $a(t_{i+1})$  is

$$p_{i+1} = \exp(f(t_{i+1}, P_i)).$$

Its quality is measured by the relative prediction error

$$e_{i+1} = (p_{i+1}/a(t_{i+1}) - 1) \cdot 100 \text{ [%]}.$$

The whole patient set is judged according to the sample mean  $m_1$  and standard deviation  $m_2$  as well as the shortest empirical confidence interval  $[l,u]$  containing 70% of relative errors.

### 5.5 Results

The results presented in the following table reflect the relevant part of experiments performed within the experimental environment new1main (C-language).

| Model type | $t_1$ [days] | No. of cases | $m_0$ [%] | $m_1$ [%] | $[l,u]$ [%] | Remark              |
|------------|--------------|--------------|-----------|-----------|-------------|---------------------|
| M1         | -            | 2175         | 30,7      | 117       | [-32,44]    | $t_1$ irrelevant    |
| M2         | 0            | 2175         | 95,5      | 235       | [-16,112]   | for comparison only |
| M2         | 1            | 1541         | 102,5     | 293       | [-44,76]    | 2/3 of cases        |
| M2         | 2            | 1177         | 46,0      | 183       | [-28,44]    | of cases            |
| M3         | -            | 2175         | 17,6      | 117       | [-36,40]    | $t_1$ irrelevant    |
| M4         | -            | 2175         | 1,2       | 102       | [-44,24]    | $t_1$ irrelevant    |

*Comparison of models described in (5.2) according to the predictive performance specified in (5.3)*

### 5.6 Discussion

The presented results are almost self-explanatory. It is worth of noticing that:

- the advocated model M1 improves estimation quality substantially,
- the standard model M2 reaches similar quality when evaluating about half of all cases only ( $t_1=2$ ),
- the biases of both compared models motivated search for alternative models M3, M4 which brought a further improvement,
- the adequate probabilistic modelling is expected to improve results by tens of percent (it was demonstrated in (19) for the standard model M2),
- the prediction horizon is often several days (for therapy); the prediction ability of the best models is excelent from this perspective.

### 6. Conclusions

In the paper, a recently proposed model describing the time evolution of cumulated activity is successfully tested on an extensive set of real data. Moreover, the tests have led to a further model improvement.

A lot remains to be done in order to get fully practicable solution. Specifically,

- full probabilistic model is to be exploited when designing parameter estimator (it will call for application of approximate multivariate integration (14));
- prior information on the parameter range has to be built-in;
- estimators have to be embedded into a new version of a routine software system JOD;
- a numerical evaluation of the cumulated activity for models M1,M3, M4 has to be prepared;
- the combined on-line (least squares) and batch (the selection of the model type) estimation technique used in the paper is worth of being developed further.

All these steps are in different stages of development and no conceptual problems are foreseen. It will need definitely a lot of effort which is, however, worth of being put in.

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## SOME RADIATION PROTECTION PROBLEMS CONNECTED WITH THE USE OF $^{186}\text{Re}$ -HEDP AND $^{153}\text{Sm}$ -EDTMP FOR PALLIATIVE THERAPY OF BONE METASTASES

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The relief of pain in patients with painful skeletal metastases, particularly in prostate and breast cancer, is usually achieved by administering  $^{89}\text{Sr}$ -chlorid (pure beta emitter with half-life of 50.5 d). Recently,  $^{186}\text{Re}$ -HEDP and  $^{153}\text{Sm}$ -EDTMP have been proposed for this purpose because these radiopharmaceuticals may be more effective than  $^{89}\text{Sr}$ -chlorid due to their capability to deliver radiation doses at higher dose rates. Patients given radioactive substances become sources of radioactivity and present a hazard to those with whom they come into contact. Departments of nuclear medicine equipped with wards can hospitalize such patients. However, inpatient admission is unpopular with patients, inconvenient for their relatives and contributes enormously to the cost of treatment. Of course, the decision whether the radiopharmaceutical administration should be done as an inpatient or outpatient procedure depends mainly on the clinical condition of a patient.

The  $^{89}\text{Sr}$  treatment can be safely administered on an outpatient basis and it would be very advantageous to administer  $^{186}\text{Re}$ -HEDP and  $^{153}\text{Sm}$ -EDTMP to ambulatory outpatients, too. So far, a little information about this problem has been published. The 6 hr isolation of patients in the department of nuclear medicine after the  $^{186}\text{Re}$ -HEDP administration is recommended by Hoekstra (1992), by Fisher (1995) and by company Mallinckrodt Medical (DiphoTher 1995). Lewington (1993) states that the  $^{153}\text{Sm}$ -EDTMP therapy necessitates hospital admission for 24 - 48 hours following injection. Havlik et al. (1995) report that the patients treated with  $^{153}\text{Sm}$  have to stay in therapy station of the department for 60 - 84 hours according to the radiation protection regulations in Austria.

The aim of our paper is to assess whether the ambulatory (outpatient) therapy with  $^{186}\text{Re}$ -HEDP and  $^{153}\text{Sm}$ -EDTMP is possible in our country.

**Physical characteristics.**  $^{186}\text{Re}$  with a relatively short physical half-life ( $T_{1/2} = 89.3$  h) has beta-emissions useful for therapy ( $E_{\text{max}} = 1.07$  MeV) and gamma-emission suitable for external imaging ( $E_{\gamma} = 137$  keV) with a photon yield of 9.45 %. Likewise,  $^{153}\text{Sm}$ -EDTMP has favourable physical characteristics - a physical half-life of 46.27 hr, 640 keV (30%), 710 keV (50 %) and 810 keV (20 %) beta emissions and 103 keV (28 %) gamma emission.

**Administered activity.** The usual activity of  $^{186}\text{Re}$ -HEDP administered to one patient ranges from 1.3 to 1.5 GBq; administered activities up to 3 GBq were reported. In case of  $^{153}\text{Sm}$ -EDTMP the administered activity ranges from 1.3 to 3 GBq, the typical one being 2 GBq.

**Biokinetics of radiopharmaceuticals.** During 3 hours after injection of  $^{186}\text{Re}$ -HEDP 20 - 40 % of activity is accumulated in bones; during 6 hours after injection 40 - 60 % of administered activity is excreted in urine.

Accumulation of  $^{153}\text{Sm}$ -EDTMP in bones is 50 - 60 % during 2 - 3 hours, the excretion in urine is 40 - 60 % during 6 hours after injection.

**Radiation protection characteristics.** Gamma-ray dose constants of  $^{186}\text{Re}$  and  $^{153}\text{Sm}$  are 4.91 and 24.4  $\mu\text{Gy}\cdot\text{m}^2/\text{hr}/\text{GBq}$ , respectively. Dose rate in 1 m distance from the adult patient administered with  $^{186}\text{Re}$ -HEDP is 1.2  $\mu\text{Sv}/\text{hr}/\text{GBq}$ , and with  $^{153}\text{Sm}$ -EDTMP 5.3  $\mu\text{Sv}/\text{hr}/\text{GBq}$  (Eary et al. 1993, Havlik et al. 1995).

According to the ICRP Publication 61 annual limit of intake (ALI) by ingestion is 20 MBq for both  $^{186}\text{Re}$  and  $^{153}\text{Sm}$ . The effective dose from  $^{186}\text{Re}$ -HEDP is 0.38 mSv/MBq (adult) and 2.6 mSv/MBq (1 - 2 year old child). The effective dose from  $^{153}\text{Sm}$ -EDTMP is 0.3 mSv/MBq (adult) and 2.1 mSv/MBq (1 - 2 year old child) (Havlik et al. 1995).

Irradiation of patient's relatives. There are two sources of irradiation of relatives (or any other individuals who spend significant time close to the patient) of patients discharged from the hospital: external exposure and internal contamination. The estimate of the effective dose to family members was made under the following conservative (worst-case) assumptions:

- radiopharmaceuticals were administered to an outpatient, i. e. the patient is discharged immediately after injection in the department of nuclear medicine;
- a family member spent time, at average distance of 1 m from the outpatient, to complete decay of radionuclide in the body; the amount of the radiopharmaceutical in the body decreased due to radioactive decay alone (biological elimination is not considered);
- ingestion by family member of 0.01 % of the excreted activity in the urine occurred during patient's stay in his family (Hoekstra 1992);

The rough estimate of the external dose received by the family member at one meter from the patient is calculated according to the following expression obtained by integrating the dose rate  $h$  ( $\mu\text{Sv/hr/GBq}$ ) to complete decay

$$D = h \times A \times 1.44 \times T_p \quad (\mu\text{Sv})$$

where  $A$  is administered activity (GBq) and  $1.44 \times T_p$  residence time (hours) of the radiopharmaceutical in the body; it is assumed that activity of radiopharmaceutical in the body decreases monoexponentially with the physical half-life  $T_p$  of the radionuclide. The administered activity is considered 2 GBq for both  $^{186}\text{Re}$ -HEDP and  $^{153}\text{Sm}$ -EDTMP. The effective dose to children can be approximately assumed to be equal to adult effective dose (Yamaguchi 1994).

In case of  $^{186}\text{Re}$ -HEDP  $h = 1.2 \mu\text{Sv/hr/GBq}$  at one meter,  $A = 2 \text{ GBq}$ ,  $T_p = 89.3 \text{ hr}$ ; then the external dose  $D = 0.31 \text{ mSv}$ .

For  $^{153}\text{Sm}$ -EDTMP  $h = 5.4 \mu\text{Sv/hr/GBq}$  at one meter,  $A = 2 \text{ GBq}$ ,  $T_p = 46.3 \text{ hr}$ ,  $D = 0.72 \text{ mSv}$ .

So far, no results of monitoring relatives of outpatients treated by these substances have been published. Some data are available on radiation doses actually received by family members of out-patients treated for thyreotoxicosis to whom  $^{131}\text{I}$  was administered. The analysis of three papers (Harbert and Wells 1974, Wasserman and Klopper 1993, Thomson et al. 1993) shows that the dose to family members ranges from 1 to 5  $\mu\text{Sv/GBq } ^{131}\text{I}$ . By multiplying the average value of 2.5  $\mu\text{Sv/GBq}$  by the ratio of gamma ray dose constants of  $^{186}\text{Re}$  and  $^{131}\text{I}$  and by the ratio of half-lives of  $^{186}\text{Re}$  and  $^{131}\text{I}$  the dose to the family member 0.11 mSv/GBq  $^{186}\text{Re}$ -HEDP is obtained. The same procedure for  $^{153}\text{Sm}$ -EDTMP yields 0.27  $\mu\text{Sv/MBq}$ . The comparison of the doses calculated by us and those estimated using measured  $^{131}\text{I}$  data suggests that our approach may be considered realistic.

The family member's ingestion of 0.01 % of the excreted activity in urine corresponds to 0.1 MBq  $^{186}\text{Re}$ -HEDP or  $^{153}\text{Sm}$ -EDTMP (it is approximately assumed that half the activity administered to a outpatient is excreted in urine); the adult's effective dose is 0.04 mSv and the 1 - 2 year old child dose is 0.26 mSv. The corresponding effective doses to family members in case of  $^{153}\text{Sm}$ -EDTMP are 0.03 mSv and 0.20 mSv.

Undoubtedly, the family member's ingestion of 0.01 % of the activity excreted in the outpatient urine is very unlikely. This statement is supported by the results of work by Buchan and Brindle (1971) who measured  $^{131}\text{I}$  uptake in the thyroid of 11 family members of  $^{131}\text{I}$  therapy patients to whom no special instructions regarding precautions had been given; the maximum uptake of 3.8 Bq per MBq administered was found. The existing information summarized in

Report NUREG-1492 (1994) suggests that internal doses to family members from intake of  $^{131}\text{I}$  are likely to be much less than external doses.

The estimates of the total exposure of family members due to  $^{186}\text{Re}$ -HEDP and  $^{153}\text{Sm}$ -EDTMP are given in Tab. 1.

Comparison with limits. In the proposed amendment of Czech regulations which will come into force in the near future it is stated that a patient containing the radioactive substance for therapeutic purposes can be discharged from the hospital if it is ensured that the annual effective dose to the family members does not exceed 5 mSv in case of adults and 1 mSv in case of children. The ICRP recommends an annual limit of 1 mSv for the general public but in special circumstances a higher value could be allowed in a single year provided that the average over 5 years does not exceed 1 mSv per year.

In Tab. 1 it is seen that the effective dose to children is not sufficiently below the annual limit of 1 mSv. It must be taken into account that the palliative therapy with  $^{186}\text{Re}$ -HEDP or  $^{153}\text{Sm}$ -EDTMP is repeated twice or more times during one year (there is minimum time interval of 6 to 8 weeks between administrations) and each injection of radiopharmaceuticals contributes to the exposure of the family members of outpatients. Also, there may be a great variation in exposure rate near patients because of a variation in retention of radiopharmaceuticals in their bodies.

Therefore, it seems reasonable to accept the 6 hr delay in a release of the patient from the hospital as it is suggested by Hoekstra (1992), by Fisher (1995) and by company Mallinckrodt Medical (DiphoTher 1995) in case of  $^{186}\text{Re}$ -HEDP. Because within 6 hours following administration about 50 % of the administered activity is excreted the doses to family members are about a half ones given in Tab.1. Moreover, patients are given instructions designed to reduce the radiation dose to family members and other members of the public by restricting the patient's behaviour.

Our calculation shows that the difference between  $^{186}\text{Re}$ -HEDP and  $^{153}\text{Sm}$ -EDTMP as to the total radiation hazard to household members is not too large. Therefore, the 6 hr stay of the patient in the department of nuclear medicine could be applied also in case of  $^{153}\text{Sm}$ -EDTMP therapy.

Conclusions. The outpatient administration of  $^{186}\text{Re}$ -HEDP and  $^{153}\text{Sm}$ -EDTMP with the subsequent keeping the patient for 6 hours in a department of nuclear medicine appears to be in compliance with regulations proposed in our country as well as with ICRP Recommendations. Of course every department of nuclear medicine planning the administration of these radiopharmaceuticals must send application to relevant regional hygienic center for a licence which must be strictly adhered to.

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**Tab. 1** Approximate estimates of the effective dose to the out-patient's family members. It assumed that a patient leaves the hospital immediately after the administration of the radiopharmaceutical.

| Substance               | Administered activity (MBq) | Effective dose (mSv/year) |      | Total effective dose (mSv/year) |
|-------------------------|-----------------------------|---------------------------|------|---------------------------------|
|                         |                             | ext.                      | int. |                                 |
| Adult                   |                             |                           |      |                                 |
| $^{186}\text{Re}$ -HEDP | 2000                        | 0,31                      | 0,04 | 0,35                            |
| $^{153}\text{Sm}$ -EDTM | 2000                        | 0,72                      | 0,03 | 0,75                            |
| 1 - 2 year old child    |                             |                           |      |                                 |
| $^{186}\text{Re}$ -HEDP | 2000                        | 0,31                      | 0,26 | 0,57                            |
| $^{153}\text{Sm}$ -EDTM | 2000                        | 0,72                      | 0,20 | 0,92                            |



## ANALYSIS OF RADIATION DOSES TO PATIENTS FROM DIAGNOSTIC DEPARTMENT OF NUCLEAR MEDICINE

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### Introduction

This study was stimulated by our interest in radiation safety of patients in nuclear medicine department. During two years (1992 - 1994) we have adopted the methodology for the determination of the collective effective dose equivalent  $S_E$  and mean effective dose equivalent  $H_E$  per exam in nuclear medicine department.

### Method

The **number of applications ( $N_R$ )** and **activities of applied radiopharmaceuticals ( $\Sigma A_R$ )** by each kind of radiopharmaceuticals and type of examinations was exactly recorded and evaluated by software developed in our laboratory. This data were used for calculation of **mean activity applied per examination ( $A_V$ )**

$$A_V = (\Sigma A_R) / N_R \quad (1)$$

Recorded data and the values of **mean effective dose equivalents per unit activity ( $H_{E/1Bq}$ )** from (2) were used for the calculation of **mean effective dose equivalent for one examination ( $H_E$ )**:

$$H_E = A_V \cdot H_{E/1Bq} \quad (2)$$

Finally the **collective effective dose equivalents** for each radiopharmaceutical and type of examination ( $S_{ER}$ ) and global **collective effective dose equivalent for department** for all radiopharmaceuticals ( $S_E$ ) during evaluated period were defined as:

$$S_{ER} = N_R \cdot H_E \quad (3)$$

$$S_E = \Sigma S_{ER} \quad (4)$$

The data for years from 1992 to 1994 were evaluated and compared with results in literature.

### Results And Discussion

1. The value of  $S_E$  (for six months period) declined during 1992-1994 from **9.01** to **5.76** man Sv, what represents decrease of about 36 %.

2. In 1993 the value was  $S_E = \mathbf{15.01}$  man Sv, what represents 14.8 % of the value of  $S_E = \mathbf{101.47}$  man Sv from all Slovak Republic (1). For the same period the number of examinations was 4985 (monthly in average  $415 \pm 48$ ), what represents 17.0 % of all examinations performed in Slovakia (29 395 exams).

3. The value of  $H_E$  was estimated to be **3.01** mSv, what represents 87.3 % of slovak mean value (3.45 mSv). These numbers are the results of lower amount of applied activities, but also by

lower contribution from the examinations required the radiopharmaceuticals with higher radiation burden.

To be aware of the world trend, we tried to replace the radiopharmaceuticals which caused relatively high radiation dose to patients ( $^{131}\text{I}$ ) with  $^{99\text{m}}\text{Tc}$ -radiopharmaceuticals (of course when it was possible). After these changes the contribution of  $^{99\text{m}}\text{Tc}$  - **labeled** radiopharmaceuticals increased from 57.7 % to 75.5 %.

4. Decreasing of value  $S_e$  (from 5.19 to 4.34 man Sv), during observed period, was mainly due to decreasing of the total number of examinations.

5. The most obvious decrease of  $S_e$  value (from 2.98 to 0.92 man Sv) was by **decreasing the contribution** from radiopharmaceuticals labeled with  $^{131}\text{I}$ . Some of them are  $^{131}\text{I}$ -OIH and  $\text{Na}^{131}\text{I}$ , which were partly replaced by  $^{99\text{m}}\text{Tc}$  radiopharmaceuticals:  $^{99\text{m}}\text{Tc}$ -MAG3 and  $^{99\text{m}}\text{TcO}_4^-$ .

The contribution from other radiopharmaceuticals labeled by  $^{32}\text{P}$ ,  $^{52}\text{Cr}$ ,  $^{67}\text{Ga}$ , or  $^{201}\text{Tl}$  varies in wide range according to requirements for these specific examinations. Their contribution to the total number of examinations and also to the value of  $S_e$  is small.

6. The monthly evaluation of  $S_e$  value is specially useful, when is for one type of examination possible to use radiopharmaceuticals of various types. The figure 1 displays diagram about the changes of number of thyroid scintigraphy and the value of  $S_e$  during year 1993 which depends on selected radiopharmaceuticals when the strategy of indication was changed. Reduction of radiation dose was done by **replacement of  $^{131}\text{I}$  with  $^{99\text{m}}\text{Tc}$  labeled radiopharmaceuticals**. We have to stress that indications for specific radiopharmaceuticals are different therefore some of them cannot be replaced by the others (causing lower radiation burden).

Our computer program enable us to compare, on monthly basis, the values of  $H_e$  and  $S_e$  and also with the mean values in previous years.

### Conclusions

The evaluation of radiation doses in nuclear medicine department is useful parameter for internal quality control. Using this method, the radiation dose in our laboratory was changed to minimum (under mean value of Slovak Republic). Unfortunately, the real data of patient's radiation doses are different from the calculated one. Due to different kinetic of radiopharmaceuticals in individual patients (influenced by pathology, age, etc.) the evaluation of radiation burden to nuclear medicine patients is problematic. But this approach enable the relative comparison of the changes in values of  $H_e$  and  $S_e$  during the observed period.

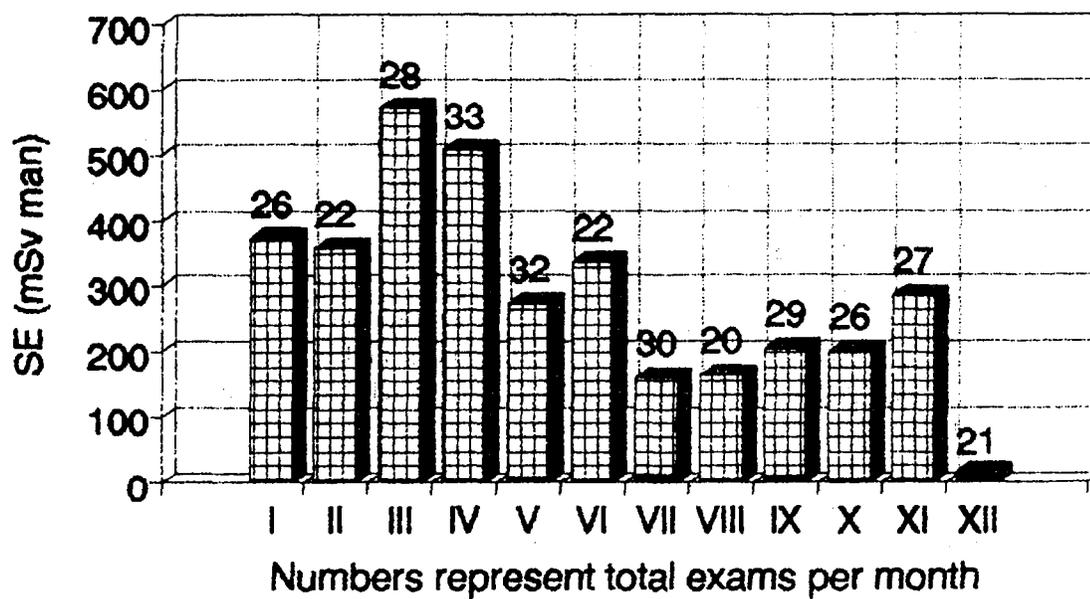
The evaluation of **Individual (minimal) effective dose equivalent - ( $H_{\text{MIN}}$ )** which represents dose calculated under physiologic conditions can be useful for indication of diagnostic examination by physicians. Therefore we propose the systematic registration of  $H_{\text{MIN}}$  from all examinations - **patient's radiation history**. This is specially important in the case of children and young people.

The importance of the proposed method, is in regulation of radiation dose from nuclear medicine diagnostic examinations, not only by the control of number and type of examinations, but also by selection of used radiopharmaceuticals and by the way how to use them.

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**Fig 1: The collective effective dose SE from thyroid scintigraphy examinations**





## THE PRESENT STATE OF BRACHYTHERAPY PRACTICE IN THE CZECH REPUBLIC

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### **Introduction**

About 40-60% of all cancer patients require treatment with ionizing irradiation during the course of their disease. Prevailing modality of the treatment is teletherapy using gamma, X and electron high energy beams.

However, in selected situations, brachytherapy represents the optimum way to deliver radiation dose with the ability to reach a close to optimal relative integral dose. New radionuclides of high specific activity correlating with high strength enable small physical size of new sources and by using appropriate small diameter flexible source carriers and advanced afterloading technology it is becoming possible to reach almost any organ within the human body. New techniques, including dose optimisation with sophisticated treatment planning and computerised afterloading systems, have revolutionised this field in the last decade.

The number of patients requiring brachytherapy is rapidly increasing every year. This is the reason why this area needs more attention and the systematic and careful analysis to prevent and detect radiation hazards in due time. There is a paucity of reliable data on the practice of brachytherapy in the Czech Republic.

### **Materials And Methods**

Population of the Czech Republic is 10 309 200 inhabitants and there are about 45 000 new cases of malignant neoplasms notified in a year.

According to the Annual Report of Czech Ministry of Health there are 29 100 patients treated for cancer in 25 radiotherapy departments. 14 of them are well equipped and 8 can be considered as on the world standard level ( i.e. they are equipped with linear accelerator, top quality dosimetry system, access to CT scanner, planning system and simulator).

Brachytherapy treatments are given in 18 of 25 radiotherapy departments.

In 1990 a Czech National Standard for Leak-Testing of Sealing Radioactive Brachytherapy Sources was prepared by Ministry of Health. As a part of this task a survey of the infrastructure and the level of radiation protection and safety in brachytherapy practice on RT departments was investigated. The results were published in Acta Hygienica in 1990.

Since 1990 the brachytherapy techniques have undergone a great development and new radionuclides (especially Cs-137 and Ir-192) and new afterloading application systems have been replacing more hazardous "classic applications" with radium tubes and needles.

In 1995 a short survey was performed to identify the development in this field. The comparison of the main features of brachytherapy practice in 1990 and 1995 is presented in Table I.

### **Discussion**

The most important change in brachytherapy practice is the progressive equipment of 6 radiotherapy departments with high dose rate remote AFL systems using Ir-192 sources. In near future three new systems will be probably installed at the departments on the "world standard level" (Ostrava, Hradec Králové, Brno) where the manual AFL (or even direct application !!) have been used up to now.

In consequence of that changes there are also changes in the number and types of applications. The number of applications was in 1994 more than twice greater than in 1990 and the number of interstitial and surface applications is progressively increasing.

The reason is:

- practically thorough radiation protection. For professionals the protection is practically of the same level as for teletherapy techniques. And
- an increasing number of indications for brachytherapy treatment, because of small dimensions of the sources which enable applications (especially interstitial), which have been completely impossible with "classical" sources.

During the last five years the amount of radium sources at the radiotherapeutical departments have been cut down significantly :

- 3 of 16 departments completely got clear of radium sources and
- at present time 13 departments is used or deposited from 20 mg to 872 mg of radium-226. (Comm.: 20 mg in one RT dept. serves as "local etalon")
- 3 departments want to dispose radium sources as soon as (financially) possible.
- 8 departments liquidated a great deal of their radium reserves. They usually saved only such amount of radium which covers the unnecessary clinical requirements (in case when they use manual AFL) or which serves as a reserve (in case when a remote AFL system is used for treatment). The amount of Ra-226 varies in such cases from 120 mg to 360 mg.

This trend should be promoted (or better -in some way financially supported) in near future to diminish the unnecessary hazard from radium sources which are often stored at the RT departments as an useless reserves.

At 1995 only one department carries out solely direct application of radium sources. All others try to perform at least some part of applications by manual AFL and they try to replace radium sources by new cesium sources. The most important finding of the 1995 survey is a fact that since 1990 six departments in Czech Republic have begun to use solely a remote AFL in clinical practice.

### **Conclusion**

In spite of the fact that radiotherapy departments in the Czech republic have been following the general European and world trends in brachytherapy techniques and that they are installing the new remote AFL systems, the reserves of radium sources are still considerably high and not all professionals carry out their applications of radioactive sources by (at least) manual AFL.

In near future the attention should be paid to the diminution of the unnecessary radium reserves at that radiotherapy departments where the small number of applications is executed or where the new remote AFL systems are used.

For professionals practically thorough radiation protection is assured when new remote AFL systems are used. But in that case an attention should be paid to the radiation safety of the patient because of the potential exposures when high dose rate systems would failed. The likelihood of this unwanted cases can only be decreased when Quality Assurance programs is introduced into practice.

**Tab. I** Brachytherapy practice in the Czech republic

| <b>NUMBER OF DEPARTMENTS : 18</b> |             |                    |             |                    |
|-----------------------------------|-------------|--------------------|-------------|--------------------|
| SOURCES                           | 1 990       |                    | 1 995       |                    |
| Type of sources                   | No. of dpt. | Activity at a dpt. | No. of dpt. | Activity at a dpt. |
| Ra<br>(tubes, needles)            | 16          | 20 - 1135<br>[mg]  | 13          | 20 - 872<br>[mg]   |
| Cs<br>(tubes, needles)            | 6           | 2 - 101<br>[GBq]   | 7           | 2 - 101<br>[GBq]   |
| Ir wires                          | 1           |                    | 1           |                    |
| Cs-AFL (LDR)                      | 2           | 28 - 56<br>[GBq]   | 5           | 28 - 86<br>[GBq]   |
| Ir-AFL (HDR)                      | -           | -                  | 5           | 370<br>[GBq]       |
| <b>APPLICATIONS</b>               |             | 1 990              | 1 995       |                    |
| <b>NUMBER of applications</b>     |             |                    |             |                    |
| In Czech republic :               |             | 1902               | 3898        |                    |
| At single depts. :                |             | 35 - 276           | 35 - 768    |                    |
| <b>TYPES of applications</b>      |             |                    |             |                    |
| Intracavitary :                   |             | 1864               | 3351        |                    |
| Interstitial :                    |             | 20                 | 37          |                    |
| Surface :                         |             | 18                 | 510         |                    |
| <b>WAY of applications</b>        |             |                    |             |                    |
| Direct appl.of sources :          |             | 2 dept.            | 1 dept.     |                    |
| Manual AFL or dir.appl :          |             | 9 dept.            | 4 dept.     |                    |
| Manual AFL (solely) :             |             | 6 dept.            | 3 dept.     |                    |
| Remote or manual AFL :            |             | 1 dept.            | 4 dept.     |                    |
| Remote AFL (solely) :             |             | -                  | 6 dept.     |                    |



## THE BEGINNING OF STUDIES AND USE OF X-RAYS IN THE CZECH LANDS

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Based on historical sources our abstract and poster aim to present the remarkably early response to Röntgen's discovery of X-rays in the Czech Lands, then part of the Austro-Hungarian Monarchy, and to call attention to some early radiological studies done here before the foundation of Czechoslovakia in 1918.

### The early X-ray experiments by physicists and engineers

Three groups of scientists started the first studies on Röntgen's X-rays at the turn of year 1895 almost simultaneously:

- physicists **Č.Strouhal** (1850-1922), **F.Novák** (1869-1944) and **O.Šulc** (1869-1901) at the Prague Czech University,
- electrical engineer **K.Domalíp** (1846-1909) and physical chemist, specialist in microphotography, **K.Kruis** (1851-1917) at the Prague Czech Technical University,
- physicist and electrical engineer **J.Puluj** (1845-1918) at the Prague German Technical University. Puluj had invented an original cathode ray tube, awarded a silver medal at the Paris international exhibition of 1881.

Reports on those physical experiments, together with demonstrations and X-ray pictures, began to appear at scientific meetings as early as from mid January 1896 and were immediately reported in newspapers and scientific periodicals (eg *Věstník České Akademie*, 4, 34, 80-89, 128-133, 281-286 (1896); *Sitzungsberichte d. k. Akademie d. Wissenschaften, Math.-naturw. Classe*, 105, 170, 228-238, 241, 243-245, Wien, 1896). X-ray demonstrations also became a popular public attraction.

### The first attempts at X-ray diagnostics and therapy

It was particularly the lecture and demonstration of X-rays by E.Strouhal at the Prague meeting of the Czech Medical Society on 17 February 1896 that arose the interest of the medical profession in the use of X-rays. A pioneer of medical roentgenology was **Rudolf Jedlička** (1869-1926). While still intern at the surgical department of the Czech Faculty of Medicine in Prague, Jedlička performed the first X-ray diagnoses using both Strouhal and Domalíp's X-ray equipments and an early X-ray apparatus bought for pleasure by a Prague hotel owner. Exposure was as long as two hours. Nevertheless the results obtained gave an impetus to installing the first X-ray equipment (manufactured by firm M. Kohl of Chemnitz, Germany) at the Czech surgical department in 1897. By 1901 its skiagram collection already numbered over 2000 negatives of interesting congenital deformities, joint inflammations and about 300 pictures of elbow injuries. Jedlička published a review of the initial results and experiences in X-ray diagnostics, inter alia, in [1, 2]. Jedlička and his co-workers also experimented with X-ray therapy. The results were reported eg at the Vth International Congress of General and Medical Electrology and Radiology held in Prague in March 1912. As Jedlička later wrote, the first biological findings had been obtained by them from histological specimens of excised skin alterations of their own hands.

**Rudolf Jaksch von Wartenhorst** (1855-1947), Professor of special medical pathology and therapy at the German Faculty of Medicine in Prague, pioneered the use of X-rays in internal medicine. The results of X-ray diagnosis of pneumonia, as obtained at his department in 1903-1905, were published eg in work [3].

R.Jaksch together with H.Přibram, J.Löwy and others were also experimenting with X-rays in the treatment of malignant tumours, aleukaemic splenomegalies and leukaemia.

Early roentgenological studies in the Czech Lands dealt also with the effect of X-ray on the eye and skin. Conducted since 1897 by **J. Chaluppecký** (1864-1918) of the ophthalmological department of the Czech Faculty of Medicine in Prague, these studies were performed initially on rabbits and then on excised porcine lenses. Chronic radiation of the rabbit's head and eye elicited alopecia and severe ulcerative alteration of the conjunctiva and cornea. According to him, X-ray caused no chemical changes in the eye lens. [4]

Application of X-ray radiation in haematology and digestive tract diagnosis was studied since 1903 at the surgical department of the above Faculty by **K.Hynek** (1879-1960). His papers published between 1905 and 1906 ranked among the first dealing with X-ray therapy of chronic and pernicious leukaemia [5]. In 1912 he extended his studies to polycythaemia as well.

In addition some experiments using X-rays in dermatology, gynaecology, dentistry, ENT and other medical disciplines in the Czech Lands date to the years before World War I.

An early summarizing chapter of X-ray therapy appeared in the monograph [6] by Czech dermatologist **E. Slavík**.

#### **The early X-ray equipment of provincial hospitals and practitioners' surgeries**

Enthusiasm on the part of individuals, together with occasional funds donated by public institutions, made it between 1896 and 1899 possible to furnish also some provincial hospitals (eg Německý Brod, Olomouc) and practitioners' surgeries with the first X-ray equipment, imported or self-assembled. Thus one of the first X-ray apparatus in Moravia was made allegedly by an enthusiast, engine operator by profession, for F.Dreuschuch, the local doctor at Náměšť nad Oslavou.

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## THE LIQUIDATION OF LIQUID RADIOACTIVE WASTE ON NUCLEAR MEDICINE DEPARTMENTS

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One of several factors which limits the service of nuclear medicine departments is the liquidation of radiation waste in accordance with environmental protection law. One can release the liquid radioactive waste to the public sewage or to the natural stream only when the following conditions are satisfied:

a) to the public sewage system

$$\sum_{i=1}^n (a_{vi}/I_{pi}) < 1 \quad (1)$$

b) to the natural stream

$$\sum_{i=1}^n (a_{vi}/I_{pi}) < 0.01 \quad (2)$$

where  $a_{vi}$ - specific activity of i-th radionuclide in waste water [Bq/m<sup>3</sup>]

$I_{pi}$ - annual limit of intake of i-th radionuclide by ingestion [Bq].

The condition a) is almost always satisfied for diagnostic nuclear medicine departments, where the dilution with non active trash is common. The waste from the therapeutical applications have to be stored temporarily in decay storages until the radioactive waste is possible to dilute. The storage time depends on the total radioactivity, the volume of waste and also on the amount of the available water for dilution. These factors determine the technical solution of storage spaces, of monitoring of specific activity and also the direct dispersal of liquid wastes.

The most serious problem for Clinic of Nuclear Medicine - NOI - St. Elis. Hosp.- Bratislava is the localization of Clinic in the downtown, inside the hospital area with the dilution water deficit. This department is the only one in Slovak Republic performing therapeutical applications.

To be able to perform the necessary amount of therapies and also to introduce a new therapeutical methods, in 1992 - 1994 the old liquidation waste disposal station /LWDS/ was reconstructed with the aim to satisfy the newest requirements of radiation hygiene.

LWDS is the 5 - floors object partly underground which satisfied the requirements for liquidation of radioactive liquid waste from diagnostic procedures (annually 5000 patients) and also from 200 therapeutical applications annually (15 beds, 720 GBq <sup>131</sup>I). The capacity of LWDS is able to store about 90 m<sup>3</sup> liquid radioactive waste.

Part of the underground spaces are used for the storage of solid radioactive trash.

The liquid waste from Clinic of NM is collected through isolated metal (rustproof, iron) sewage system to the storage with continuous observation of water specific activity. According to the activity, the liquid waste is placed to the 5 decay storages with the volume about 15 m<sup>3</sup>. The sixth one serves for the case of technical accident. When the activity declines, the liquid waste is diluted with non active medical trash to the level which is acceptable by law about radiation hygiene protection. The storage walls are made from Ba - concrete 25 - 50 cm thick which is enough for sufficient protection of operation staff and also for walking around persons.

Double-layer high quality chemical material prevents the water leak and diffusion of radionuclides into the concrete.

Technology consists of cast-iron drains, powerfull slush pumps, operation valves, regulation technology from dosimetric system for continuous monitoring of specific activity , for managing system with powerfull industrial computer.



## DEPOSITION OF $^{60}\text{Co}$ AND $^{137}\text{Cs}$ ADSORBED ON ZEOLITE IN MATRICES ON THE BLAST FURNACE SLAG

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The cement matrices on Portland cements have been found appropriate for an immobilisation of the low and medium level radioactive wastes. The matrices on blast furnace slag basis are able to avoid of decreasing of strength of cement casts even in very inconvenient composition of waste waters. Generally, the double step procedures consisting in sorption of radionuclides to suitable inorganic or organic carriers and their incorporation into cement casts as fillers are seemed to be preferred. The experimental data (leaching tests and compressive strength measurements) are presented for case the radionuclides from model water solution and radioactive waste water were uptake on natural and chemically modified granular zeolitic tuffite which show improved sorption ability and were subsequently incorporated into cement casts on blast furnace slags (BSF) basis. The milled, previously granulated blast furnace slag from Iron Works Košice, Slovak Republic, was used as the principal matrix former in all experiments. The amounts of the water glass activator ( $S_m = \text{SiO}_2/\text{Na}_2\text{O} = 1.9$ ;  $r = 1.37 \text{ g.cm}^{-3}$ ) corresponded 3.7 wt.% of  $\text{Na}_2\text{O}$  of dry weight of BSF. The all experiments were carried out with zeolitic tuffite from Nižný Hrabovec, Slovak Republic.

The natural zeolite was treated by the NaOH solution of the following concentrations: 1M, 2M, 4M or 6M respectively. The blends of individual components were casted into rectangular prism moulds (1x1x3 cm). The cast consisted invariably of 90 % of BSF, 10 % of fine zeolite and solution of water glass and to this basic mixture were added either 10 or 20 % granular (grob) active, or non active zeolite during the mix formation, in excess to content of previous solid components. The water/cement ratio varied in interval 0.32-0.40. Model solutions labelled by  $^{137}\text{Cs}$  or  $^{60}\text{Co}$  have been used for sorption experiment and real waste water concentrate from Nuclear power plant Jaslovské Bohunice as well. The real water consisted of  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ , in borate and nitrate form.

The radioactivity of dry cement casts was measured in 4 geometry. The leaching experiments were performed in three solutions: water (W), 0.03 M HCl (A) and 0.04 M NaOH (B).

The mechanical properties of sample with non active granular zeolites and their corrosion characteristics as reflected by the bulk density changes, after placement of samples in liquid media, are given in Table 1.

The mechanical strength of samples decreased in a line: non filled matrix, matrix filled with natural zeolite, with zeolite treated by 1M, 4M and 6M NaOH solution respectively. Increased portion of zeolite decreases the mechanical strength of samples. After 42 days of exposition in the corrosive liquid the mechanical strength of samples did not decrease at all with an exception of 20 % of 4M zeolite kept in the acid solution. In majority of cases the strength was equal, or higher than the 28 days strength of samples prior to their corrosive tests. The results are summarised in Table 2. The experiments showed that leaching of radionuclides, which were sorbed from model solution and radioactive waste water, in water and bases solution were negligible. Fig. 1 - 2 show changes of radioactivity of casts, prepared from model solution, in acid solution, which were less than 5 %, and Fig.3 shows the changes of radioactivity of the casts, prepared from waste water, in acid solution as well. The mechanical strength of prepared composites is favourable and shows good compatibility of zeolitic additives with a basic matrix. Leaching experiments and mechanical strength are hopeful and show good retention of observed radionuclides in samples exposed in leaching solutions.

**Tab. 1** Bulk densities and the strengths of specimens

| Sample    | Bulk density [g.cm <sup>-3</sup> ] |      | Bending strength [MPa] |      | Compress. strength [MPa] |      | Porosity [vol.%] |
|-----------|------------------------------------|------|------------------------|------|--------------------------|------|------------------|
|           | 7 d                                | 28 d | 7 d                    | 28 d | 7 d                      | 28 d |                  |
| No addit. | 2,11                               | 2,12 | 3,0                    | 12,0 | 48                       | 80   | 3,8              |
| N - 10%   | 2,07                               | 2,08 | 2,2                    | 6,7  | 41                       | 68   | 6,1              |
| N - 20%   | 2,01                               | 2,05 | 2,7                    | 6,0  | 45                       | 65   | 6,4              |
| 1M - 10%  | 2,06                               | 2,08 | 1,9                    | 10,4 | 43                       | 70   |                  |
| 1M - 20%  | 2,02                               | 2,04 | 1,6                    | 8,9  | 34                       | 58   |                  |
| 4M - 10%  | 1,96                               | 1,97 | 1,4                    | 5,0  | 26                       | 40   |                  |
| 4M - 20%  | 1,83                               | 1,86 | 0,8                    | 3,6  | 16                       | 30   | 7,4              |
| 6M - 10%  | 1,96                               | 1,98 | 3,2                    | 7,7  | 30                       | 43   |                  |
| 6M - 20%  | 1,89                               | 1,91 | 1,1                    | 3,3  | 26                       | 36   |                  |

**Tab. 2** Mechanical strength of samples exposed in the corrosive medium after 42 days

| Granular zeolite |                | Compressive strength [MPa] |    |     |
|------------------|----------------|----------------------------|----|-----|
| Sample           | Content [wt.%] | Corrosive solution         |    |     |
|                  |                | W                          | B  | A   |
|                  | 0              | 93                         | 90 | 108 |
| N                | 10             | 78                         | 80 | 72  |
| N                | 20             | 61                         | 69 | 68  |
| 1M               | 10             | 66                         | 83 | 72  |
| 1M               | 20             | 69                         | 68 | 61  |
| 4M               | 10             | 59                         | 55 | 48  |
| 4M               | 20             | 39                         | 33 | 24  |
| 6M               | 10             | 57                         | 57 | 57  |
| 6M               | 20             | 40                         | 44 | 38  |

**Fig. 1** The leachability of radionuclides from the casts with addition of 10 % zeolite adsorbent (model solution) in acid solution; - + - natural zeolite, - . - zeolite modified by 1M NaOH, - o - zeolite modified by 4M NaOH, - ■ - zeolite modified by 6M NaOH

**Fig. 2** The leachability of radionuclides from the casts with addition of 20 % zeolite adsorbent (model solution) in acid solution; - + - natural zeolite, - . - zeolite modified by 1M NaOH, - o - zeolite modified by 4M NaOH, - ■ - zeolite modified by 6M NaOH

**Fig. 3** The leachability of radionuclides from the casts with addition of zeolite adsorbent (waste water) in acid solution; - o - addition of 10 % zeolite adsorbent, - ■ - addition of 20 % zeolite adsorbent

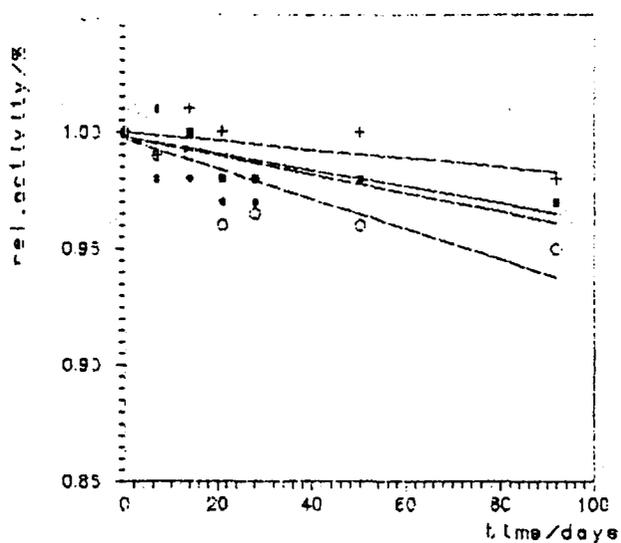


Fig. 1

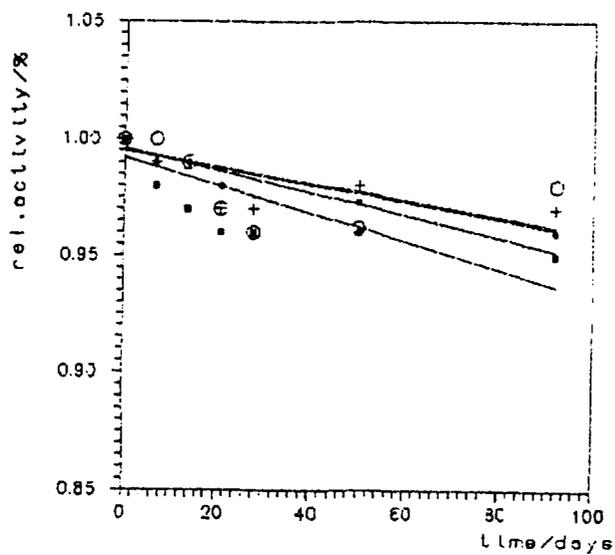


Fig. 2

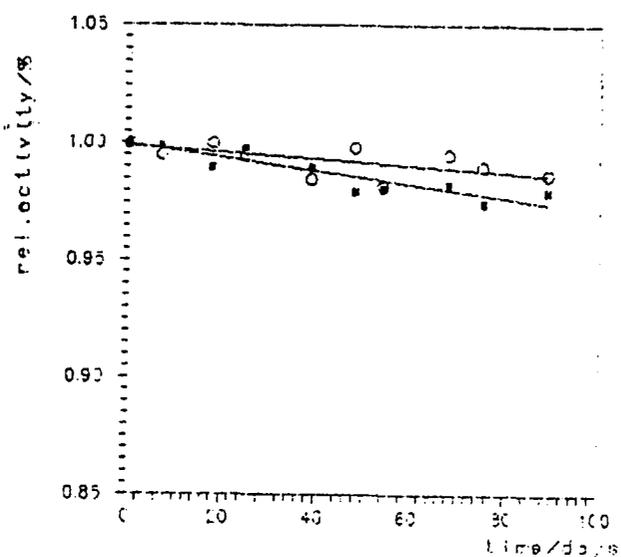


Fig. 3



## MEASUREMENT ON THERAPEUTICS SIMULATOR XIMATRON

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### **Abstract**

This year we commemorate the 100<sup>th</sup> anniversary of the discovery of X-rays. During these 100 years the use of X-rays in medicine underwent great progress both in the field of technical development and protection of medical staff and patients. In September 1995 the latest model of simulator of the firm Varian Ximatron has been installed at the National Cancer Institute in Bratislava. The present communication reports on some hygienic measurements carried out at this Institute which can provide insight in the present state of modern equipments.



## LIMITATION OF RADIOACTIVE DISCHARGES FROM NPP BASED ON RADIONUCLIDE SPECIFIC MONITORING

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### **Summary**

Monitoring of nuclear power plants (NPP) discharges into atmosphere and hydrosphere based on particulate-iodine-gase (PIG) measurements and gross beta or gamma plus tritium measurements respectively is being improved by nuclide specific measurements employing semiconductor gamma and alfa spectrometry and radiochemical methods. In connection with this progress new concept of authorized effluent limits and the related regulations is being implemented in the Czech Republic. Activities of all principally contributing to the effective dose radionuclides discharged during a year multiplied by a Sv/Bq conversion coefficient based on a standard model are summed up and the effective dose is compared with the new limit. These limits should be authorized for discharges into atmosphere as per caput collective committed effective dose in the NPP surrounding or as its value multiplied by the number of surrounding segments providing thus committed effective dose to member of a virtual critical group and into the hydrosphere as committed effective dose to the critical group member. The criteria for obligatory evaluation of the specific radionuclide contribution and the resulting requirements on minimum detectable activities for both the shorter and longer monitoring intervals are included in the new regulations. The P-I-G and gross activity measurement results will be compared with investigation levels only and no more used for limitation purposes.



## FUEL LEAK TESTING PERFORMANCE AT NPP JASLOVSKÉ BOHUNICE

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### Introduction

The fuel cladding is, after the fuel pellet matrix itself, the second and, from the view point of normal operation, the most significant barrier preventing the release of fission products into the reactor coolant. Regular and proper leak testing of fuel assemblies is an inevitable requirement closely related not only to operational regulations but also to an attempt to assure optimal operation from the radiation protection point of view.

### Leak testing performance at NPP Bohunice

Until 1986 all fuel sipping tests were performed by using Soviet wet canister-pool system. During refuelling outage several fuel assemblies were tested. This type of leak testing equipment was not optimal, mainly because of the long testing time.

In 1986 an in-core sipping equipment designed and manufactured by Siemens KWU was purchased [1,2]. Since then, full core sipping tests were performed in all cases, if the leak occurrence has been proved by operational data. This decision about sipping test was based on primary coolant activity measurements during the reactor operation and iodine spike analyse.

The extent to which fission products are released from the fuel rods to the coolant inside the channel is dependent on the temperature increase of the fuel. During the leak testing the flow of coolant through the assembly is therefore throttled by blocking its upper end. The decay heat produces a temperature rise in the fuel with a subsequent increase in the fission gas release. The temperature effect at the sipping tests causes the rise of free gas volume (up to 3%).

In a defective rod, the fission gases expand and escape through the leak into the surrounding water. The resulting increase of fission products activity can be measured in water samples taken from the assembly. These samples are analysed using a germanium detector.

The nuclides caesium 134 and 137, iodine 131 and xenon 133 are used for identifying leaking assemblies. These fission products are either produced directly during reactor operation or as daughter nuclides of short lived fission products. Their half-lives are sufficiently long to be detected shortly after reactor shut-down or even after long storage periods (via caesium isotopes).

The NPP Bohunice in-core sipping equipment was also used once at the NPP Paks in Hungary (1991) and twice at NPP Dukovany (1991,1992) in Czech Republic.

In total 24 full core tests were performed at 7 VVER-440 units till March 1994 [3,6].

### Occurrence of leaking fuel assemblies and probable causes of leaks

Based on NPP Bohunice in-core sipping tests results 32 leaking fuel assemblies were identified (see table 1).

Tab. 1 Occurrence of leaking fuel assemblies in NPP Bohunice

|                                   | Unit 1 | Unit 2 | Unit 3 | Unit 4 |
|-----------------------------------|--------|--------|--------|--------|
| Number of leaking fuel assemblies | 9      | 22     | -      | 1      |

The time dependence of 31 leaking fuel assemblies at the first two units of NPP Bohunice (V-1) is shown in the Fig.1a and Fig.1b.

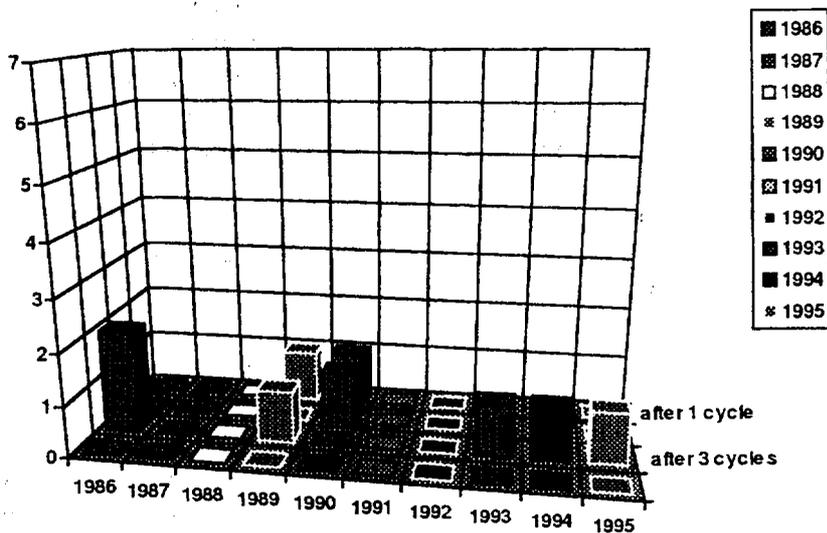


Fig.1a Time dependence of damaged FAs at unit 1 of the NPP V-1

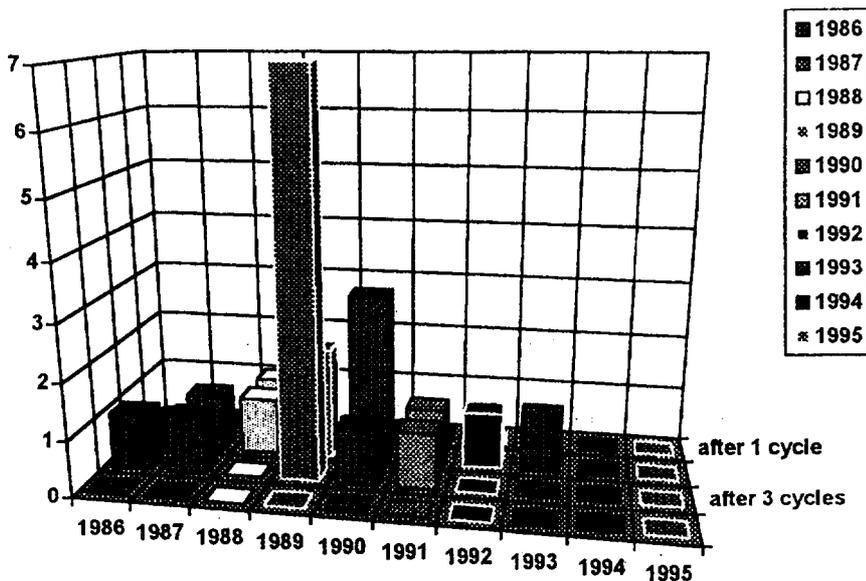


Fig.1b Time dependence of damaged FAs at unit 2 of the NPP V-1

VVER-440 core has a hexagonal symmetry. One sixth of the core matrix was used to display the last in-core position of all leaking fuel assemblies. This analyse shows that 31% of all leaking fuel assemblies found were placed directly in the neighbourhood of the power controlling assemblies. This fraction is unexpectedly high in comparison to the fraction of all fuel assemblies in their neighbourhood which is 13,4% in a full core loading pattern and 15,2% in a reduced core loading pattern (In 1985 36 dummies were placed at NPP Bohunice unit 2 at the core periphery). This fact shows that PCI mechanism should take part in the leak origin, however a set of 32 fuel leaks is statistically too small for making any serious conclusion. Also Russian reports and experience as well as experience obtained at NPP Bohunice units 3 and 4 do not assume high probability of PCI mechanism.

Another interesting fact is the high number of leaking fuel assemblies at Bohunice unit 2 (total 22). Because of large leak number in one cycle with fading in subsequent cycles, debris induced fretting as a leak cause can be assumed. After subtracting of cycles 1989 and 1990 is unit Bohunice 2 comparable to unit Bohunice 1 or Loviisa 1, 2. In spite of this we can assume that the leak occurrence at NPP type V-230 is more frequent than at NPP type V-213 (presented by Bohunice 3,4, Dukovany 1,2,3,4 and Paks 1,2,3,4).

All these identified leaking fuel assemblies were tested in the Russian canister pool sipping system as well. The results of these tests showed that also the Russian canister-pool sipping system is able to discover leaks.

Some tests showed, that in case of small leaks, the sensitivity of both methods was comparable, while for large cladding defects canister-pool sipping provides higher sensitivity. The main disadvantage of canister-pool sipping method at VVER-440 is its time demand, which does not allow full core testing during refuelling. The necessary time for canister-pool sipping is 14 days, while for in core sipping is only 48 hours. However, according our experience the in core sipping method is nowadays the proper way for useful leak testing management from operational point of view.

### **The mast sipping method**

Recently we have still been working at the mast sipping method for VVER-440 leak tightness control. On the basis of experiences with in-core sipping the in-mast sipping method for VVER-440 fuel assemblies was developed. The equipment is prepared for manufacturing and in connection with the well proven Siemens control panel can bring new benefit in fuel testing. The advantages of mast sipping method are :

- the leak testing is made during the fuel handling time,
- the leak testing and the fuel transport to the spent fuel pool are made by one grip of fuel assemblies.

The first advantage saves the time, which was necessary before for the leak testing and the second advantage increases the safety for operating staff and equipment by reduced volume of handling with the refuelling machine [5,7].

The mast sipping method used beside temperature also the pressure effect for leak testing, which is more effective. By pressure effect the fission products are released from fuel rods due to the change in the hydrostatic pressure as the assembly is moved vertically upwards during fuel handling operations. This rise in pressure in the case of VVER-440 reactors is up to  $10^5$  Pa.

The mast of the VVER-440 handling machine is modified for the sipping process by the installation of a movable hood at the end of the gripper. As an assembly is gripped and lifted during a normal fuel handling operation, the hood slides down, by its own weight, to a position at the upper end of the fuel, interrupting coolant flow through the assembly. The assembly is then transferred to the spent fuel pool while the leak testing is conducted simultaneously. The hood returns to its normal position on contact with surrounding assemblies the moment it is

released. The movable hood is held in both position by an inflatable seal. Inside the gripper, bleed lines are installed, which are routed through the mast wall to the testing equipment. For the testing of water and gas samples, an existing Siemens control panel is possible to use at Bohunice.

### **Conclusion**

The NPP Bohunice VVER-440 fuel leak testing experience are relatively extensive in comparison with other VVER-440 users. As the first East Europe NPP we adapted Siemens (KWU) in core-sipping equipment to VVER-440 units and since this time we have done these tests also for NPP Paks (Hungary) and NPP Dukovany (Czech Republic).

The occurrence of leaking fuel assemblies in NPP Bohunice is in the last 5 years relatively stabilised and low. A significant difference can be observed between type V-230 (31 leaks) and type V-213 (1 leak).

None of the indicated leaking fuel assemblies has been investigated in the hot cell. Therefore we cannot confirm the effective causes of leak occurrence. Nevertheless, the fuel failure rate and the performance of leak testing in NPP Bohunice are comparable to the world standard at PWR\*s.

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## **AUTOMATION OF METROLOGIC OPERATIONS ON MEASURING APPARATUSES OF RADIATION MONITORING SYSTEM**

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*NPP Dukovany*

The primary question of operation of nuclear power installations is their safety and minimal influence over the environment. The system of metrologic care about the measuring apparatuses of ionizing radiation is an important item of the radiation monitoring of NPP Dukovany operation.

The radiation monitoring of the operation is of a rather wide scope. It includes monitoring of persons, space inside as well as outside of the nuclear power plant, technology, gaseous and liquid releases, and the like. The measuring apparatuses of ionizing radiation may be, in the basic division, divided into three groups: measuring apparatuses of the central information and measurement system of radiation monitoring, measuring apparatuses of autonomous measuring circuits and portable measuring apparatuses used by the personnel.

The system of metrologic care about the measuring apparatuses of ionizing radiation makes possible calibration and verification of these measuring apparatuses. The establishment of a metrologic laboratory for measuring apparatuses of ionizing radiation was given already in the design of NPP Dukovany. Laboratory operation, i.e. uniformity and correctness of metrologic performance in the first place, is ensured by a three-men staff consisting of two college graduates and one high-school graduate. Since 1990 the laboratory is accredited and then licensed for metrologic operation on measuring apparatuses of the so called controlled sphere. It has the statute of a state metrologic centre in the conformance with the Metrology Law.

The laboratory ensures by its own resources the metrology of especially:

- film and thermoluminescent dosimeters
- ionization chambers and Geiger-Müller counters
- scintillation and semiconductor detectors
- neutron dosimeters and detectors.

In 1994 over 2000 metrologic operations were done which represents, in comparison with 1990, increase by almost 61%. Measuring apparatuses are treated metrologically in regular, usually one-year, intervals and always after a repair. The number of measuring apparatuses repairs used in the ten-year-operated nuclear power plant has not an increasing tendency and has no influence over the increasing number of metrologic performances. About 100 of different types of mostly Czech, Russian, German and French produced measuring apparatuses are included in the collection of operated equipment. In addition, about 150 given measuring apparatuses from the state controlled sphere are verified in the laboratory during one year.

The rather great number of metrologic operations must not affect the resulting work quality, of course. The questions of quality are of the first place importance and conform to the provisions of European standards of EN 45000 and ISO 9000 series. The laboratory is subjected to frequent internal and external inspections, checkings by the workers of state supervisory bodies - State Office for Nuclear Safety, Czech Metrology Institute - Ionizing Radiation Inspection and Public Health Office.

The increase of metrologic operations number has been made possible only by a timely reconstruction of the laboratory and by computerization of the measuring procedure and of administrative work which consists mainly of recording of a great number of information pieces about the observed measuring apparatuses.

There are three working places in the laboratory:

- irradiation gamma stand with Cs-137 sources
- irradiation stand with Pu-Be neutron sources
- spectrometric working place.

Work with irradiation stands might be observed by means of a closed TV circuit. Automation of the measuring procedure is conditioned by an active application of a bus bar, which is designated as IEEE-488, IEC-625, GP-IB and sometimes also HP-IB. There are at disposal in the laboratory measuring apparatuses which might be, by means of the above-mentioned bus bar, connected into a measuring system directed by a personal computer. We succeeded in fully automating the management of the irradiation stand - the selection of single standard sources. Automatic setting of the position of the optical testing bench cart belongs among highly successful circuits; on the cart the checked detector of ionizing radiation is placed. The position is measured by an incremental sensor in the range of 0.6 to 7.2 m from the irradiation stand. The following evaluation circuit of our own construction, based on the use of so-called Wilkes circuit, with control storage of the ROM type makes possible to reach the accuracy of position measurement up to 0.1 mm. The computer sets the cart position with the accuracy up to 1 mm, which is sufficient for practical operation. During a metrologic operation the detector is placed into a beam of ionizing radiation, its type is selected on the screen of a personal computer and what remains is a question of computer art and measuring technique. An algorithm is worked out for each type of the detector - a methodic procedure of calibration or verification in conformance with which the whole metrologic operation is done as for programme.

With regard to the uniqueness of the laboratory operation, all the work in the sphere of hardware as well as software has been implemented by our own forces. The equipment of the laboratory makes possible to test metrologically all the radiation monitoring apparatuses used in NPP Dukovany. The quality of operation of the laboratory of ionizing radiation metrology qualifies the proper functioning of the radiation monitoring system, which directly influences the ensurance of nuclear safety of NPP Dukovany.



## SELECTIVE NOBLE GASES MONITOR

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The monitoring of real releases from a ventilation stack of VVER NPP requires a system by several orders more sensitive than currently used radiometer Kalina, designed to cover the range up to a design-based accident. To reach this goal a noble gases monitor with a germanium detector (MPVG) has been developed. It enables nuclide selective monitoring of current value of volume activity of particular nuclides in ventilation stack and daily releases of noble gases (balancing).

MPVG can be viewed as a system build of three levels of subsystem:

- measuring level
- control level
- presentation level

Measuring level consists of gamma-spectrometry system and operational parameters monitoring unit (flow rate, temperature, humidity)

Control level provides communication between presentation and measuring level, acquisition of operational parameters and power supply.

The presentation level of MPVG enables:

- the measured data storage in predetermined time intervals
- the presentation of measured and evaluated values of radiation characteristics.

The interconnection of different MPVG levels is ensured by serial data links.

The performance of an experimental MPVG based on GEM-10175-P-Plus detector with relative efficiency 10 % and resolution 1,75 keV is characterized by:

Limit of detection for 24 hours balancing of releases:

- 1200 Bq/m<sup>3</sup> for <sup>133</sup>Xe (without a cartridge)
- 1 to 3 Bq/m<sup>3</sup> for <sup>133</sup>Xe (with a cartridge)

Limit of detection for monitoring with 60 min. response time:

- 5,9 kBq/m<sup>3</sup> for <sup>133</sup>Xe (without a cartridge)
- 5-15 Bq/m<sup>3</sup> for <sup>133</sup>Xe (with a cartridge)

The range of volume activities monitoring with inserted sorbent cartridge:

from 10 to 10<sup>6</sup> Bq/m<sup>3</sup> (for <sup>133</sup>Xe)

The range of volume activities monitoring without sorbent cartridge:

from 10<sup>4</sup> to 10<sup>9</sup> Bq/m<sup>3</sup> (for <sup>133</sup>Xe)

The monitored radionuclides - default set:

<sup>41</sup>Ar, <sup>85m</sup>Kr, <sup>87</sup>Kr, <sup>88</sup>Kr, <sup>89</sup>Kr, <sup>131m</sup>Xe,  
<sup>133</sup>Xe, <sup>133m</sup>Xe, <sup>135</sup>Xe, <sup>135m</sup>Xe, <sup>137</sup>Xe, <sup>138</sup>Xe.

(Modification is possible.)

Basic time interval for monitored data update:

- configurable in minutes

Time interval for balancing releases:

- the balancing is performed for the previous day

Data storage (archive generation):

- volume activities for monitored radionuclides for last 720 measurements
- balancing data for the last 365 days.

The prototype monitor has been operated during the whole 1993. The stability of the preset parameters made any resetting needless. The monitored data have been transformed into weekly histograms of selected radionuclides volume activity and submitted to the operator (EBO).

During the whole monitor testing period the noble gases releases have been extremely low - well below the limit of detection for 1 hour measurements except for  $^{135}\text{Xe}$ . The volume activity of  $^{133}\text{Xe}$  only occasionally exceeded detection limit  $10 \text{ Bq/m}^3$  while those for  $^{135}\text{Xe}$  varied between 10 to  $50 \text{ Bq/m}^3$ . Values for  $^{41}\text{Ar}$  were from below detection limit ( $1000 \text{ Bq/m}^3$ ) up to  $2000 \text{ Bq/m}^3$ .

The values of volume activities observed at maximum releases have been approximately ten times higher. In that case in balancing some other nuclides exceeded corresponding detection limits:

$^{88}\text{Kr}$  (67 22)  $\text{Bq/m}^3$

$^{85\text{m}}\text{Kr}$  (17 7)  $\text{Bq/m}^3$

$^{135\text{m}}\text{Xe}$  (7.1 0.5)  $\text{Bq/m}^3$

$^{138}\text{Xe}$  (5.9 0.9)  $\text{Bq/m}^3$



## PLANNING FOR ENVIRONMENTAL RESTORATION OF THE CONTAMINATED BANKS NEAR NPP BOHUNICE

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### Summary

The 18 km long banks of the Bohunice NPP waste water recipient are contaminated by <sup>137</sup>Cs as a result of two accidents on the CO<sub>2</sub> cooled NPP-A1 unit in 1976 and 1977. Since 1992, all the contaminated waste waters dumping from NPP Bohunice has been carried out directly to the Váh River through a specially constructed 15 km long pipeline.

Radiological surveys and detailed monitoring of this contaminated site was carried out in the period of 1991-1994 y-s. Radiological characteristics on the canal and Dudváh River bank surfaces are given detailly in an earlier paper [1]. In average, 6.3 Bq <sup>137</sup>Cs/g of soil was identified on the 18 km long affected bank surfaces. The 10 typical sections was identified depending on the characteristic contamination and type of the banks, as it is marked in Tab.1. In the Váh River banks, where the monitoring was finished lastly, in 1994, the contamination was identified only in a few flood plain sections with overall length of about 1km, close to the present Dudváh River mouth (Vxi in Tab.1).

Thus, data in Tab.1 represent the final extent of contamination in the Bohunice site. The overall contaminated area in this site with <sup>137</sup>Cs activity above 1 Bq/g of soil is about 67000 m<sup>2</sup> and thus, the corresponding volume of top 20 cm thick soil layer is about 13000 m<sup>3</sup>.

After finalisation of the monitoring works, in 1994, it was recognized that due to the very low acceptance limit (1 Bq/g) [2] and the capacity reasons it is practically impossible to ensure safe disposal of the resulting volume of contaminated soil inside the Bohunice NPP area, which is the only acceptable place in the site. To be able to propose an acceptable and rational scope of the contaminated banks restoration a considerable re-considering of the previously used approach and planning of these measures was necessary to carry out.

Environmental restoration belong to a costly intensive remedial measures. Choice of appropriate techniques, and also, the derivation of proper acceptance limits depend on approach choosen (recovery or planned activities) and a number of hardly obtainable data (basic dose limit, parameters of the selected radiation risk scenarios...) that necessarily should be harmonized and clearly declared beforehand by the competent authorities.

A clear legislation is absent in this field. It was thus necessary, first of all, to develop certain principles and rules for evaluation of the necessary extent of remedial measures, including derivation of contamination acceptance and cleanup limits, and to achieve their authorization by the hygiene authorities. The principles, as well as the

evaluation itself were recently elaborated in the VÚJE Institute.

Selection of critical radiation risk scenario, harmonisation of their parameters and ICRP recommended limitations for recovery on the level of 1 mSv/y of individual effective dose enabled the authors to develop proper site specific derived acceptance and cleanup limits. Appropriate bank restoration technologies were proposed [3], as well, taking into account identified type of the contaminated banks [1,2] and the costs and benefits achieved. According to comments of the Institute of Preventive and Clinical Medicine (ÚPKM Bratislava), the submitted principles and derived criteria [4] were approved by the State Health Institute in Bratislava in the beginning of 1995y. Goal of the presentation is to describe the mentioned Principles developed, criteria derived, as well as the present stage of planning for the contaminated banks restoration.

Two stay scenarios were selected for the evaluation of actual risk on the banks and contaminated field, and another two residential scenarios for the evaluation of a potential risk from the use of contaminated soil supposed to be fully (about 200m<sup>3</sup>) and partially (about 50 m<sup>3</sup>) spread on the site surface around a resident's living house and in his garden.

The presence and future residual contamination on the banks were considered as a source under control within the time period of 50 years (NPP vicinity). However, the "contaminated soil use" type of (residential) scenario can not be considered as improbable. Typical parameters [5] for a partial and full residential scenario, considering no dilution of top soil from the banks, were utilised as potentially the most critical one for proper acceptance and cleanup limits derivation (see Tab.2). Exceeding of the limiting dose for these scenario, only justifies implementation of the cost consuming restoration techniques.

For optimizing less costly remedial measures (warning signs...) an agreed scenario with a pre-estimated factor for collective dose (milk + E<sub>ext</sub> from banks)  $2 \times 10^{-7}$  man Sv.y<sup>-1</sup> / (m<sup>2</sup>.Bq<sup>137</sup>Cs.g<sup>-1</sup>) was applied. Limitation of individual effective doses according to a site specific bank stay scenario was also considered for this purposes with a limiting value of 0.25 mSv/y.

Cost analysis of available remedial techniques were carried out, too. Two techniques have been selected for the contaminated banks restoration project:

1. removing/disposal of 20 cm soil top layer from steep and unengineered banks, and
2. mechanical dilution/fixation of contamination by clean 15 cm soil cover for the contaminated flat areas (terraces of the engineered part of Dudváh River banks).

Two-fold reduction of the anticipated potential radiation risk (comparing with the not-deluted contaminated soil use) were accepted, maximally, for the lastly mentioned technique, however cost saving is considerable (about 10-time lower the cost comparing to removing/disposal technique one).

According to the dose factors in Tab.2 (50 m<sup>3</sup> and 200 m<sup>3</sup> of soil use) and Principles developed, and assuming 25 cm thick and 2.5m wide strip of contaminated top soil layer on the banks, the basic acceptance limits AL for <sup>137</sup>Cs in soil and critical size of continuously contaminated bank areas were derived as:

AL<sub>200</sub> = 6.0 Bq/g and 800 m<sup>2</sup> (300 m) or AL<sub>50</sub> = 8.0 Bq/g and 200 m<sup>2</sup> (80 m) for removing/disposal of the soil on steep unengineered banks. For clean soil covering technique, on the Dudváh flat terraces, the resulting limits are, consequently, in an

interval  $AL_{50C} = 8$  upto 16 Bq/g. For warning sign application on the banks:  $AL_{ws} = 4$  Bq/g on 2500 m<sup>2</sup> of continuously contaminated area or 1 km of bank. For removing of the contaminated spots (canal Manivier) with a small area,  $AL_{25} = 25$  Bq/g (area cca 1 m<sup>2</sup>) was approved according to the proposed exemption levels in [5].

According to the criteria developed, it is necessary to subject to restoration about 11 000 m<sup>2</sup> of contaminated area on the Dudváh River banks and 8000 m<sup>2</sup> on the Manivier canal banks (Tab.1). On the Dudváh R., most of this area are in the engineered section DR3 where clean soil cover on 9500 m<sup>2</sup> of contaminated flat terraces is sufficient to be applied in accordance with the accepted principles. On the canal banks, only, the spots of contamination proposed to be removed because off soil disposal capacity saving.

On Dudváh River, due to the strongly non-uniform top soil activity distribution, it is sufficient to apply the cleanup criterion  $AL_{50} = 8$  Bq/g, only (Tab.1). The proper residual activities will thus automatically comply the more severe limit  $AL_{200}$  for some larger area of the cleaned-up banks. This cleanup criterion seems to be optimal, also, from the point of view of top soil <sup>137</sup>Cs activities volume distribution on the contaminated banks.

Thus, the re-evaluated extent of the banks restoration should include removing and safe burial of about 1100 m<sup>3</sup> of contaminated soil from and covering by clean 15 cm soil layer on about 10 000 m<sup>2</sup> of the banks. Current techniques used for maintenance and engineering of the water bodies are planned to use at these restoration works. The only acceptable place for the storage facility with regard to the public opinion expressed by the mayors of villages is the site of the Bohunice Plant. The contaminated soil have to be buried in a subsurface isolated disposal facility.

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Tab.1 Contaminated areas and  $^{137}\text{Cs}$  activity concentrations on the affected banks near Bohunice NPP

| Contam. section | S, > 1            | As     | S, > 8            | As     | A-resid | Note   |
|-----------------|-------------------|--------|-------------------|--------|---------|--|
|                 | [m <sup>2</sup> ] | [Bq/g] | [m <sup>2</sup> ] | [Bq/g] | [Bq/g]  |  |
| K1              | 10000             | 6.7    | 2000              | 9.5    | 4.9     | strip, 1-1.5m<br>strip, 0-2.5m<br>(near village) |
| K2              | 5730              | 16.2   | 5730              | 16.2   | 3.3     |  |
| K3              | 9725              | 2      | 0                 |        | 2       |  |
| D1              | 1200              | 1.8    | 0.0               | 0.0    | 1.8     | (land field)                                     |
| D2              | 1500              | 3.5    | 0.0               | 0.0    | 3.5     |  |
| D3              | 3725              | 4.7    | 1400.0            | 8.4    | 2.5     |  |
| Dpo             | 1500              | 2      | 0.0               | 0.0    | 2.0     |  |
| DR1             | 5940              | 1.9    | 0.0               | 0.0    | 1.9     | soil covering                                    |
| DR2             | 6050              | 3.2    | 0.0               | 0.0    | 3.2     |  |
| DR3             | 10500             | 9.6    | 9450.0            | 10.2   | 0.9     |  |
| VPK1            | 4760              | 2.1    | 10.0              | 8.7    | 2.1     | S < min area                                     |
| VK35            | 2530              | 2.8    | 25.0              | 9.2    | 2.8     | S < min area                                     |
| VK6             | 4260              | 1.8    | 0                 |        | 1.8     |  |
| SUM             | 67420             |        | 18615.0           | 0.0    | 2.      |  |

Note: S,>1,>8 - area with activity conc. >1,>8 Bq/g

Tab.2 Dose factors (DF) related to 1 Bq/g of  $^{137}\text{Cs}$  in soil and DILs for selected criterial scenarios

| SCENARIO                          | To [y]  | geom.f            | t <sub>exp</sub> [h/y] | INGESTION [REL.UNIT] | DF [mSv/y] | DIL(1,T <sub>0</sub> ) [Bq/g] |
|-----------------------------------|---------|-------------------|------------------------|----------------------|------------|-------------------------------|
| STAY ON BANKS                     | 0       | 300x1.4<br>g=0.54 |                        | 0.4<br>milk+meat     | 0.035      | 28.6                          |
| STAY ON CONT.FIELD                | 0       | 500<br>g=0.67     |                        | 1<br>veg.+potato     | 0.078      | 12.8                          |
| USE OF SOIL<br>50 m <sup>3</sup>  | 5<br>0  | 1950<br>g=0.39    |                        | 1.2<br>ve+po+mi+me   | 0.14       | 8<br>7.1                      |
| USE OF SOIL<br>200 m <sup>3</sup> | 10<br>0 | 2000<br>g=0.67    |                        | 1.2<br>ve+po+mi+me   | 0.21       | 6<br>4.8                      |

Rel. unit of ingestion = 0.04 mSv/y (potato 110 kg + root veg. 55kg + leaves veg. 55kg)  
 $g = (\text{used dose rate}/\text{Bq}\cdot\text{g}^{-1}) / (0.118 \text{ microSv}\cdot\text{h}^{-1}/\text{Bq}\cdot\text{g}^{-1})$  (geom. f. against a half-indefinite source)  
 ve - vegetable, po - potato, mi - milk, me - meat

$\text{DIL}(1, T_0) = (1/\text{DF}) \cdot \exp(\lambda T_0)$ , where  $T_0$  is time from which the scenario likelyhood is considered as non-zero



## TRITIUM VOLUME ACTIVITY IN NATURAL WATERS OF NPP TEMELÍN REGION

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After setting in operation, the nuclear power plant Temelín (NPPT) will become an important source of tritium contamination of the local environment [1]. This was the reason for starting the tritium monitoring network in 1991 with the aim to determine a long-term trend and the range of tritium background level fluctuations before NPPT start-up. In the frame of this network the samples of river water, air humidity and precipitation are collected in the vicinity of NPPT as well as in several more distant points and analysed for tritium (HTO) content [2].

This paper presents the results of tritium measurement in selected rivers of NPPT region obtained during the period 1991-1994.

### **Sampling and Measurement**

Particular attention is paid to Vltava river into which liquid effluents will be discharged and which is also utilized as a drinking water supply for the capital Prague. Samples from the Vltava river were collected downstream near the mouth of NPP waste canal (point Hladná) and in front of the intake into Prague water works (point Podolí). Tritium content was analysed also in surface waters of Palečkův, Temelínský and Strouha streams which can be affected by gaseous effluents due to atmospheric removal processes. Sampling was performed by standard procedure in the monthly intervals. Details of sample preparation as well as description of sampling sites was published in [3].

Tritium activity was measured with Tri-Carb 1050 TR/LL liquid scintillation counter under optimised conditions [4] enabling its content to be determined with relative error 10-20% on the  $1\sigma$  significance level.

### **Results and Discussion**

The time courses of tritium concentrations in an individual water streams are shown in Figs 1-2. The annual averages along with maximum and minimum values in the corresponding data sets are presented in Table 1.

The comparison with the similar data of foreign laboratories shows that results of our measurements approach those which are characteristic for the continental climatic zone of Central Europe. As apparent from Figs 1 and 2, no extreme values exceeding the obvious range of natural tritium fluctuation have been recorded: this indicates that so far no other source of this radionuclide could interfere with the NPPT future emissions in this region. The time series of all presented tritium data exhibit the seasonal variations with the pronounced summer maximum which is consistent with the stratospheric origin of this radionuclide.

Table 1 shows that mean annual tritium activities of investigated river waters varied within 1.9-3.0 Bq/l during the period 1991-1994 and that their trend has been slowly decreasing. This fact, as well as the seasonal variability, suggests, that tritium level in the surface waters of studied region is largely governed by this radionuclide global atmospheric fallout.

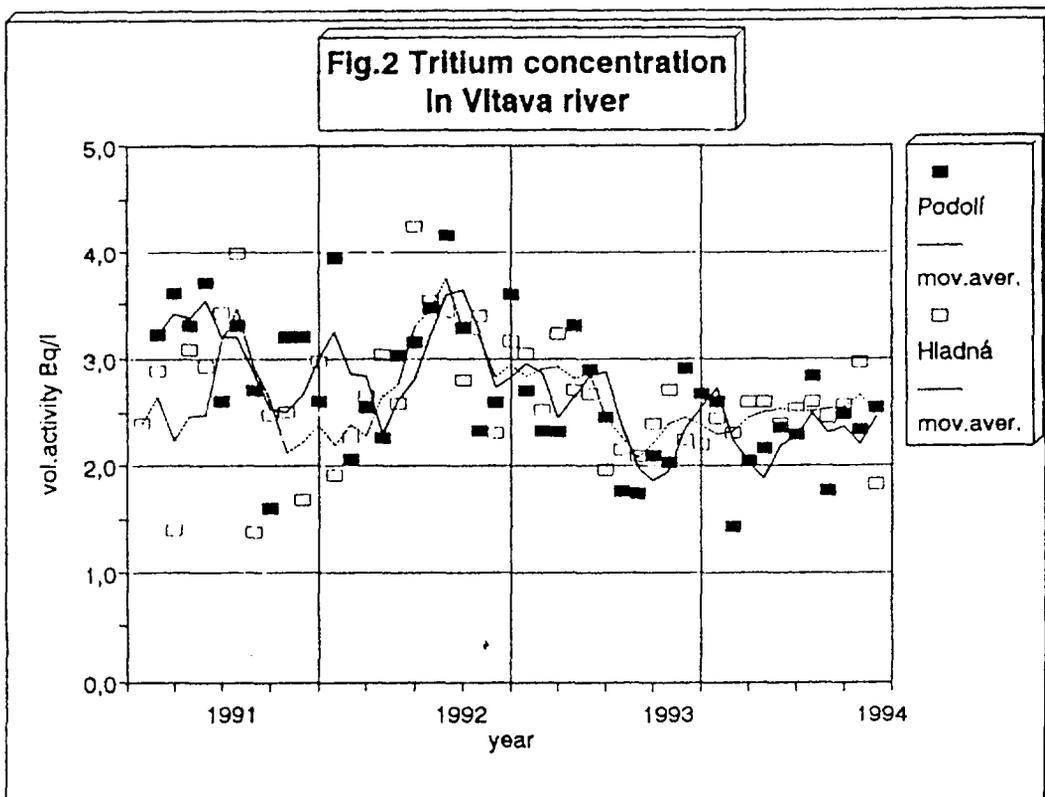
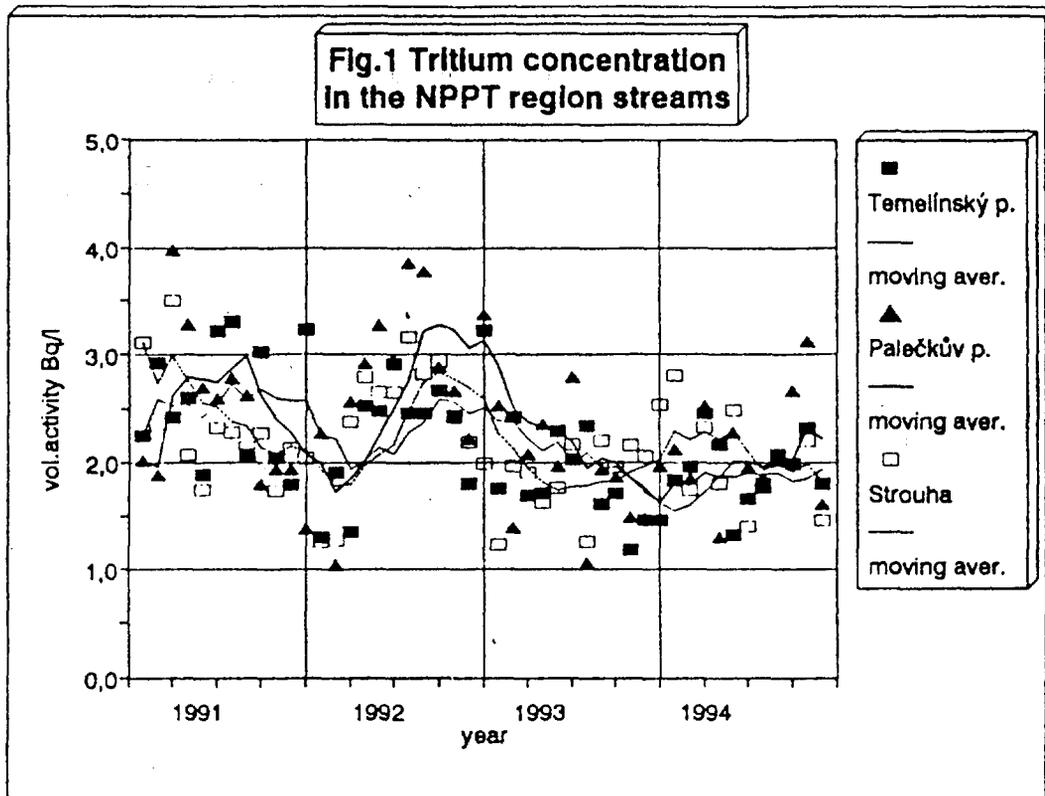
The results of this work indicate the trend of background tritium in examined natural waters and make possible the evaluation of their potential future contamination.

**Tab. 1** Average and extreme tritium concentrations [Bq/l] in river waters of NPPT region.

| Sampling station | Vltava<br>Hladná Podolí |        | Temelín.<br>potok | Palečk.<br>potok | Strouha |
|------------------|-------------------------|--------|-------------------|------------------|---------|
| 1991             | 2.6                     | 3.0    | 2.5               | 2.6              | 2.3     |
| 1992             | 2.9                     | 3.0    | 2.3               | 2.6              | 2.4     |
| 1993             | 2.6                     | 2.5    | 2.0               | 2.0              | 1.9     |
| 1994             | 2.4                     | 2.3    | 1.9               | 2.1              | 2.0     |
| 1991-4           | 2.6                     | 2.7    | 2.2               | 2.3              | 2.1     |
| min. 1991-4      | 1.4                     | 1.4    | 1.2               | 1.0              | 1.2     |
|                  | (9/91)                  | (3/94) | (11/93)           | (3/93)           | (2/93)  |
| max. 1994-4      | 4.3                     | 4.2    | 3.3               | 4.0              | 3.5     |
|                  | (7/92)                  | (9/92) | (8/91)            | (3/91)           | (4/91)  |

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## SELECTIVE LEACHING OF AEROSOL PARTICLES COLLECTED BY CASCADE IMPACTOR IN THE VENTILATION STACK OF NPP V1 IN JASLOVSKÉ BOHUNICE

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### Summary

The study was a part of investigation of the size distribution of aerosol in air effluents from NPP V1 Jaslovské Bohunice. We tried to evaluate the possible relationship between aerodynamic diameter of aerosol particles and chemical forms of radionuclides attached to the discharged aerosol. Selective leaching was used for speciation of radionuclides present in the aerosol particles and for the estimation of their behaviour in the environment and absorption in gastro-intestinal tract.

Two parallel sets of samples were collected by means of two six-stage cascade impactors placed into the ventilation stack of the NPP V1. Activity concentrations of the radionuclides in the air, collected on collection substrates taken from individual impactor stages and on back-up filter, were determined by sensitive gamma spectrometric analysis using HPGe detectors. In the experiment, the collected aerosol was divided into four groups. Approximate aerodynamic diameter intervals of aerosol particles within the groups and aerodynamic diameter cut-off for the cascade impactor stages are given in Tab.1. For the individual groups seven leaching steps were used. The composition of the solutions used is given in Tab.2. The successive leaching steps are so ordered that any subsequent leaching should leach fractions solubilized in preceding leaching step, too. The leaching by the third solution should correspond to the gastric medium.

We determined following 12 radionuclides:  $^{110m}\text{Ag}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{54}\text{Mn}$ ,  $^{103}\text{Ru}$ ,  $^{124}\text{Sb}$ ,  $^{125}\text{Sb}$ ,  $^{113}\text{Sn}$ ,  $^{65}\text{Zn}$ ,  $^{95}\text{Zr}$ . No essential differences were found between parallel samples. We, therefore, evaluated every pair of samples together, using mean values of activities. The mean values of total activity concentration (the sums of mean activity concentration of the six stages and the back-up filter) are shown in Fig.1. Our results show that the leached fraction of the activity concentration does not depend on the size of the aerosol particles. The leached fractions of individual radionuclides at 7 leaching steps and the cumulative leached fractions are shown in Fig.2 and Fig.3. The most easily leached radionuclides are isotopes of cesium, silver and ruthenium, the least leachable being tin and zirconium. The cumulative fractions for the third and seventh leaching steps are given in Tab.3. The differences in the leached fractions between isotopes of the same elements are supposed to be caused by the measurement error. The first leached fraction (see Fig.1) might be underestimated whilst the second fraction may be overestimated due to possible sorption on collection substrate, because no carriers were added to leaching solutions.

We would like to continue the experiments in future, collecting samples under different conditions of reactor operation. The speciation will be done for integral samples (without size distribution) for the sake of attaining reasonable measuring times.

Acknowledgement: We thank I.Hýbela from NPP Jaslovské Bohunice for operation and control of cascade impactors.

**Tab. 1** Characteristic of the 4 groups of aerosol and of the cascade impactor

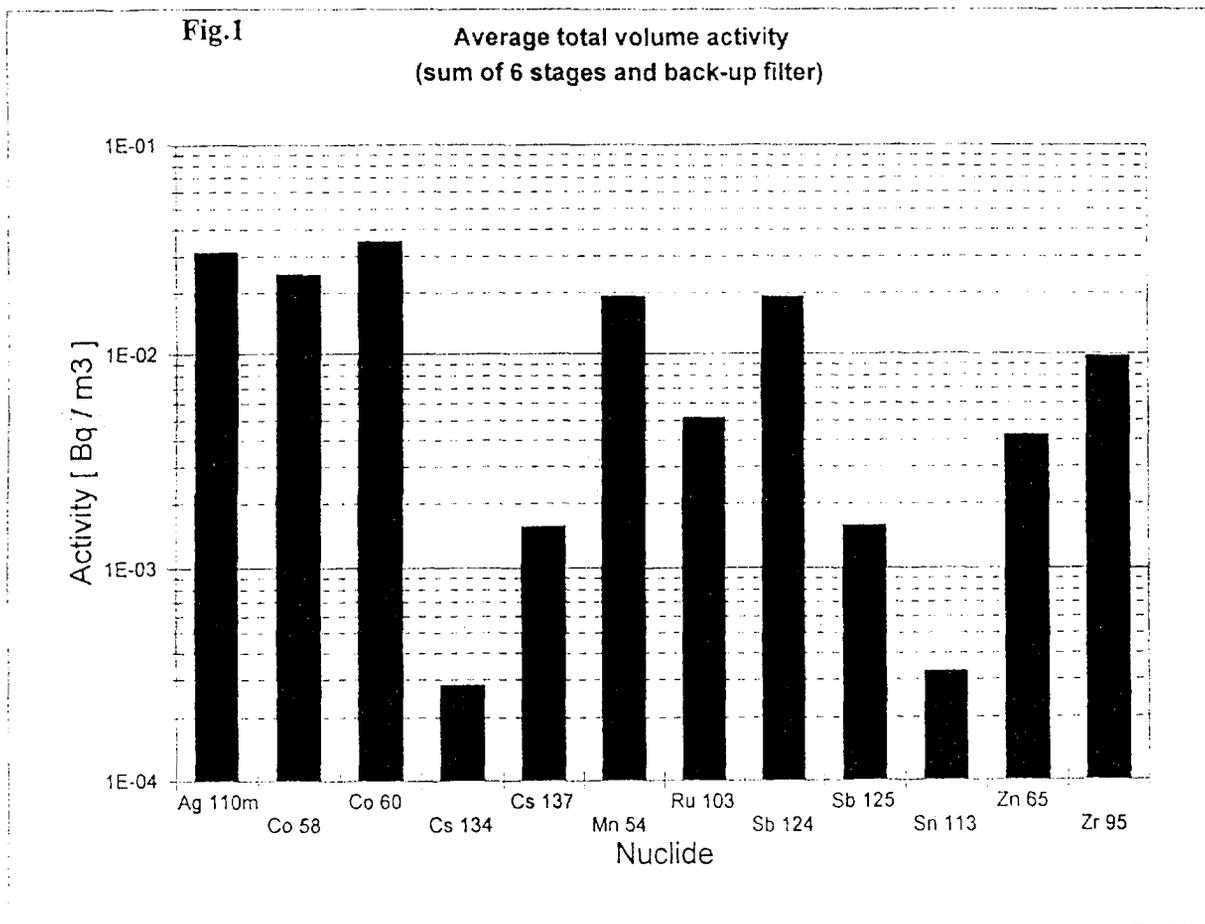
| Group number | Aerodynamic diameter interval of aerosols in group<br>[ $\mu\text{m}$ ] | Impactor stage number | Aerodynamic diameter cut-off<br>[ $\mu\text{m}$ ] |
|--------------|---|-----------------------|---|
| 1            | > 10.2  | 1                     | 10,2  |
| 2            | 1.3 - 10.2  | 2                     | 4,2   |
|              |   | 3                     | 2,1   |
|              |   | 4                     | 1,3   |
| 3            | 0.39 - 1.3  | 5                     | 0,69  |
|              |   | 6                     | 0,39  |
| 4            | < 0.39  | back-up filter        | approx. 0.01                                      |

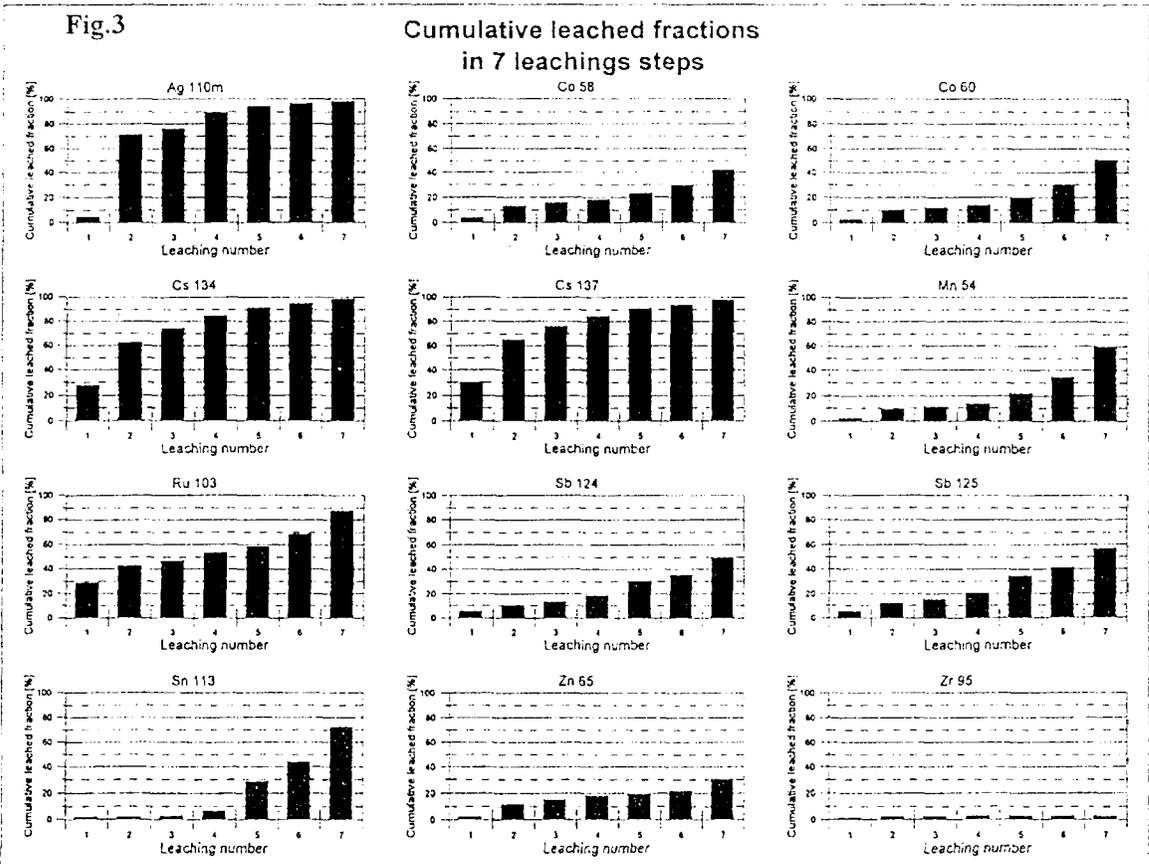
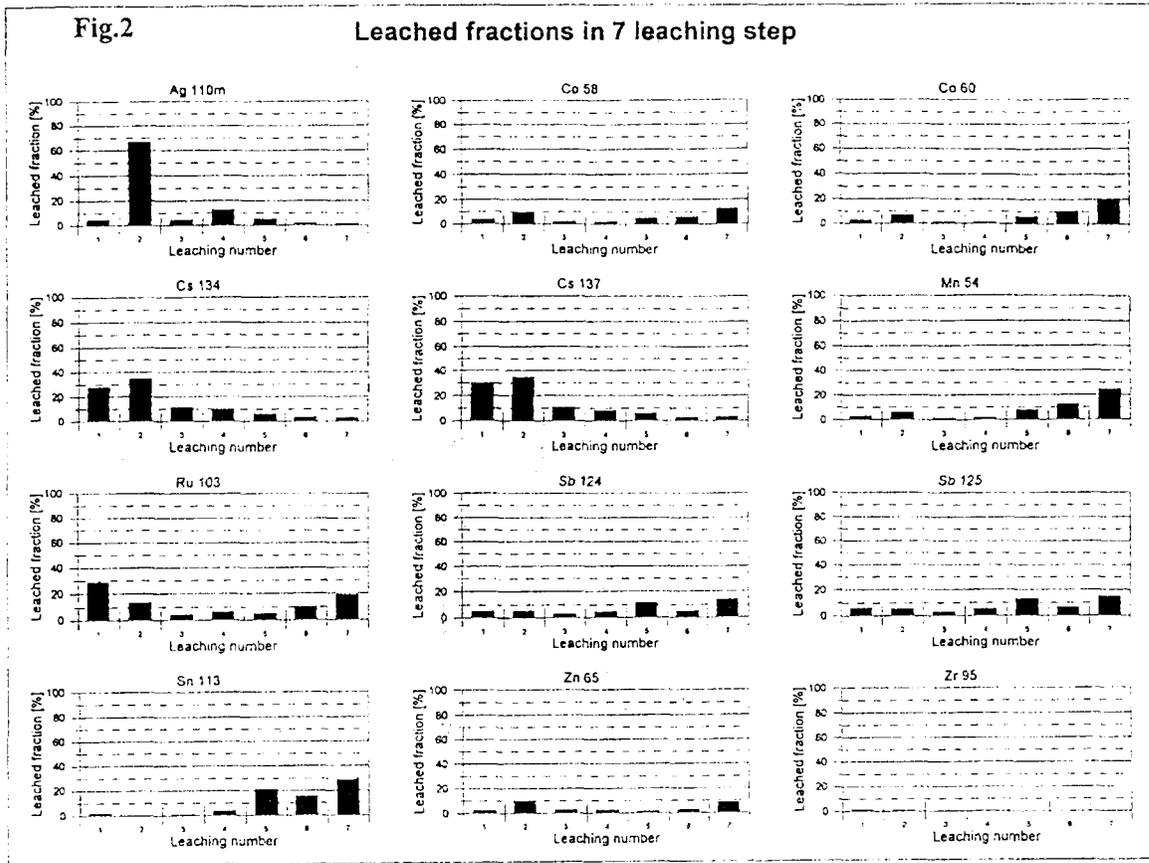
**Tab. 2** Leaching solutions

| Leaching number | Leaching solution  |
|-----------------|--|
| 1               | distilled water - water soluble part   |
| 2               | 1M $\text{MgCl}_2$ in 0.5M( $\text{NH}_4\text{CH}_3\text{COO} + \text{NaCH}_3\text{COO}$ ) at pH 7 - exchangeable ions |
| 3               | 0.1M HCl - modelling of the stomach solutions  |
| 4               | 0.5M $\text{NH}_2\text{OH}\cdot\text{HCl}$ in 25% $\text{CH}_3\text{COOH}$ - leaching of iron and manganese oxides     |
| 5               | 6M HCl - metals bound in insoluble compounds   |
| 6               | 7M $\text{HNO}_3$ - organically bound metals   |
| 7               | 7M $\text{HNO}_3$ warm leaching - the rest of the organically bound metals   |

**Tab. 3** Cumulative leached fraction after the 3rd and the 7th leaching step

| After the 3rd leaching    |               | After the 7th leaching    |               |
|---------------------------|---------------|---------------------------|---------------|
| Nuclide                   | Fraction in % | Nuclide                   | Fraction in % |
| $^{110\text{m}}\text{Ag}$ | 77            | $^{134}\text{Cs}$         | 99            |
| $^{137}\text{Cs}$         | 76            | $^{110\text{m}}\text{Ag}$ | 98            |
| $^{134}\text{Cs}$         | 75            | $^{137}\text{Cs}$         | 98            |
| $^{103}\text{Ru}$         | 47            | $^{103}\text{Ru}$         | 88            |
| $^{58}\text{Co}$          | 16            | $^{113}\text{Sn}$         | 73            |
| $^{125}\text{Sb}$         | 15            | $^{54}\text{Mn}$          | 60            |
| $^{65}\text{Zn}$          | 15            | $^{125}\text{Sb}$         | 57            |
| $^{124}\text{Sb}$         | 14            | $^{60}\text{Co}$          | 51            |
| $^{60}\text{Co}$          | 12            | $^{124}\text{Sb}$         | 50            |
| $^{54}\text{Mn}$          | 11            | $^{58}\text{Co}$          | 43            |
| $^{113}\text{Sn}$         | 3             | $^{65}\text{Zn}$          | 31            |
| $^{95}\text{Zr}$          | 3             | $^{95}\text{Zr}$          | 3             |







## UPTAKE OF RADIONUCLIDES OF CAESIUM AND COBALT ON MODIFIED ZEOLITE AND THEIRS DEPOSITION IN MATRICES ON BLAST FURNACE SLAG

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One of the possibility for radioactive waste liquid immobilisation is the incorporation of radionuclides onto ion exchangers. From this point of view the natural zeolite are very promising because they are cheap and their radiation stability is high. The sorption ability of zeolite is very good for some metals Ag, Cu, Pb and Cs, but on the other hand, it is negligible for Co. The zeolite sorption ability can be improve by chemical treatment. The main idea of all experiments was to find a way to increase the sorption ability of zeolite for Cs and Co. By means of chemical treatment amonium, potassium, sodium and H<sup>+</sup> - form of zeolite were prepared. The chemical modifications of zeolite were carried out with

- 2M solution of NaNO<sub>3</sub>, NH<sub>4</sub>NO<sub>3</sub>, KNO<sub>3</sub>
- 0.1 M solution of HCl
- NaOH solution of different concentration.

The methode of model radioactive solution was used to find the sorption ability for Cs and Co of every modified zeolite mentioned above. The model solution were 0.05 M solution of Co labeled by <sup>60</sup>Co or Cs labeled by <sup>137</sup>Cs. The highest sorption ability was observed for zeolite modified by NaOH. This was the reason why further detailed experiments on the influence on the influence of NaOH on zeolite sorption ability were done. In these experiments 0.16 M, 0.5 M, 1.5M, 2 M, 4 M and 6 M NaOH modifying solution were used. In order to determine the influence of competitivw ions K<sup>+</sup>, Na<sup>+</sup>, Ca<sup>2+</sup>, H<sup>+</sup> and Cs<sup>+</sup> or Co<sup>2+</sup>, the distribution coefficients ( $K_d$ ) of Co and Cs were measured as a function of competitive ion concentration. These experiments show that  $K_d$  decrease from low to higher concentration of competitive ions. It is interesting, the influence of Cs<sup>+</sup> ions on sorption of Co is negligible, although the excellent sorption ability of zeolite for Cs is well known. Probably the uptake of Co and Cs are of different type and therefore Cs and Co are not competitive ions.

The influence of pH on uptake of Cs and Co by modified zeolite was searched as well. The variation of  $K_d$  of Cs with the pH for modified zeolites is very similar to that for natural zeolite. The dependence of  $K_d$  for Co on pH is also very similar to that for natural zeolite but only up to pH 7. At this point is a dramatic decrease of  $K_d$ . This result, together with the incompetitive uptake of cobalt and caesium, shows the uptake of Co is not an exchange reaction but some type of chemisorption. On the other hand, the variation of distribution coefficient with concentration of competitive ions shows that modified zeolite by NaOH are excellent material for the simultaneous uptake of cobalt and caesium.

The very important chatacteristic of sorbent material is the leachibility in natural enviroment. The experiment showed that in acid solution the leachibility of Co from modified zeolite is not adequately low. But in combination with appropriate matrix this drawback is suppressed. The cement matrices on Portland cements have been found appropriate for zeolite to immobilisate Co and Cs in low and medium level radioactive wastes. The matrices on blast furnace slag basis are able to avoid of decreasing of strength of cement casts even in very inconvenient composition of waste waters. Generally, it seems that this double step procedure

consisting in uptake of radionuclides to zeolite and its incorporation into cement casts to be preferred.

The experimental data (leaching tests, compressive strength measurement and porosity) were measured for the case the Co and Cs from model water solution and radioactive waste water were uptaken on chemically modified zeolite and were subsequently incorporated into cement casts on blast furnace cement slags (BSF) basis (Iron Works Košice, Slovak Republic). The activator was water glass in amount corresponding to 3.7 wt.% of Na<sub>2</sub>O of dry weight of BSF. The compressive strength and porosity showed decreasing with amount of added zeolite and with degree of treatment of zeolite, but in every case was satisfactorily high, for both model water solution and waste water. The leachability was tested in water, in basic solution and in acid solution. The leachability in water and basic solution was negligible, in acid solution it was less than 4 % which is inside of value of applied measure method.

The compressive strength, porosity and leaching experiment are hopeful and show good mechanical stability and good retention of observed radionuclides in samples exposed in leaching solutions.