IDENTIFICATION AND ASSESSMENT OF HIGH RADON AREAS IN SOME RURAL REGIONS OF THE BALKANS

Doctoral Dissertation
Jožef Stefan International Postgraduate School
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IDENTIFICATION AND ASSESSMENT OF HIGH RADON AREAS IN SOME RURAL REGIONS OF THE BALKANS

Doctoral Dissertation

IDENTIFIKACIJA IN OCENA OBMOČIJ Z VISOKIMI KONCENTRACIJAMI RADONA V NEKATERIH PODEŽELSKIH DELIH BALKANA

Doktorska disertacija

Supervisor: Prof. Dr. James P Mc Laughlin

Co-Supervisor: Prof. Dr. Peter Stegnar

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Extended Abstract

The thesis deals with a specific aspect of a general survey, that is being carried out during last ten years in several regions of Serbia (former Yugoslavia, former Serbia and Montenegro) to assess population exposure to natural radioactivity based on geochemical and integrative pattern research approach. The originality regarding this work is related to the facts such as follows:

- the *first identification and assessment* of high areas of natural radiation in Serbia which provides insight into its regional characteristics,
- the *interpretation of the results in terms of geological aspects, building types and human habits,*
- the *first introduction and field applicability* of both (surface and volume trap) *retro techniques* in Serbia and
- *assessment of doses and risks* to the population in investigated high natural radiation rural communities.

Introduction

Although radon, being the most important natural source of population radiation exposure, has been a well explored issue for Serbia there is no systematic data on the indoor radon exposure in most Balkan countries. As reliable assessment of population exposure to natural radiation environment requires proper structured program of radiation measurements and due to the fact that the program should be based on thorough analysis of background information on geology of local territory, building construction traditions and population lifestyle, several hundred houses in rural communities of South Serbia and Kosovo, Southeast and West Serbia, in Republic Srpska (Bosnia and Herzegovina) and in Montenegro has been investigated since 1997. From the geological point of view each region has its own specific peculiarity and some of are associated with high level of natural radiation; e.g. Niška Banja, Southeast Serbia - presence of radium rich travertine; Gornja Stubla - combined uranium-thorium anomalous zone and Kalna, Southeast Serbia - former uranium mining area in granite massive. Protocols of the field investigations to assess population exposure in these regions include indoor and outdoor radon measurements, air kerma measurements as well as soil and water radioactivity measurements. The attention was paid mainly to indoor radon exposure. Various advanced techniques of indoor radon measurements were applied involving both surface and volume retrospective techniques simultaneously at the same indoor spaces. Indoor thoron gas concentrations were investigated as well. Performed analysis of radon surveys data included study of seasonal and long-term variation, comparison of contemporary and retrospective measurements, analysis of factors affecting indoor radon levels, dose and risk assessment.

Material and methods

Contemporary radon and thoron measurements

Several differently designed chambers such as: SSI/NRPB detectors, the CR-39 detectors enclosed in small cylindrical (5 cm height, 3 cm diameter) diffusion chamber, passive discriminative CR-39 Radopot and Raduet detectors [Tokonami et al., 2005], passive discriminative polycarbonate UFO detectors [Doi et al., 1994; Žunić et al., 2003] were used in this study. Exposure periods were generally 3 months covering one complete season. Annual averages were obtained using either results of all the seasonal measurements, if available, or results of some periods corrected with seasonal factors [Žunić et al., 2007]. The exposed polycarbonate films were etched electrochemically [Žunić et al., 2003].

Retrospective Assessment of Radon Exposure (RARE) techniques

In recent years, two methods of retrospective radon assessment have been developed. Both the methods are based on measuring the activity of long-lived radon daughter ^{210}Pb. The ^{210}Pb has been accumulated by alpha particle recoil implantation over many years in the surface of solid media (mostly glass), known as surface traps (ST) [Samuelsson, 1988; Samuelsson et al., 1992; Lively and Steck, 1993; Falk et al., 1996], or in the bulk of porous media (mostly furniture filling sponges), known as volume traps (VT) [Oberstedt and Vanmarcke, 1996]. Activity of ^{210}Po can be related to the long-term average indoor radon concentration. While the ST technique is based on solid state detectors (CR-39 and LR115), the VT technique uses radiochemical procedures and alpha spectrometry. Both retro techniques, i.e., ST and VT were applied successfully in three rural communities: Kalna, Gornja Stubla and Niška Banja. These techniques have been tested for a large scale surveys and indoor spaces data were collected by surface traps, volume traps and contemporary radon measurement techniques [Žunić et al., 1999; Paridaens et al., 2000] as well as a study by Paridaens et al. [2001].

In the ST technique, glass usually found in photograph and picture frames is used, because it is often easy to date. The measurement material for the VT technique is typically a 100 cm^3 polyester foam sample taken from mattresses or cushions used in a dwelling. One of the major disadvantages is that the technique is destructive and so occupants are sometimes reluctant to supply samples for the measurement. In principle the measured long lived radon progeny should be directly proportional to the long term average radon in the house and unlike the ST technique is independent of aerosol conditions in room air.

Placement and retrieval of detectors in dwellings

The fieldwork has been carried out by the members of the Institute of Nuclear Sciences “Vinča” (Belgrade). A detailed questionnaire was completed for each room of interest that recorded a number of other relevant factors relating to indoor radon. These factors were then used in conjunction with the modified Jacobi room model [Jacobi, 1972] in order to better estimate the average radon concentration. The ST monitors were left in-situ for a period of approximately 3 months before being removed and sent to Ireland (University College Dublin) for analysis. The VT samples were sent to Belgium (SCK·CEN, Mol) for analysis.

Long-term changes of indoor radon concentration

When comparing the retrospective and contemporary measurements results it is necessary to take into account the long-term indoor radon changes [Yarmoshenko et al., 2005]. Changes and transformations of construction components (such as basement, construction joints, insulation, interface gaps etc.), building underlying soil physical condition and occupant living habits may in the course of time result in systematic changes of the radon entry. Analysis of retrospective indoor radon concentrations estimated over different ages of volume or surface traps can be somehow biased under condition of substantial long-term change of indoor radon. For example retrospective estimates of indoor radon concentration in the same space using two separate objects of different age may be found to be inconsistent. On the other hand the difference between radon concentrations determined by retrospective and contemporary technique can be rather explained by long-term changes than by strong measurements error. Thus the consideration of such processes allows meaningful comparison of contemporary and retrospective data as well as investigation of its pattern using these data. By the results of radon entry modeling the long-term character of the indoor radon variation is close to linear [Yarmoshenko et al., 2005] though the year-by-year variation could be random. In particular, to describe monotonous long-term changes a coefficient k equal to the ratio of the annual radon concentration of two consecutive years has been introduced. Using this coefficient, retrospective indoor radon concentration CR (average of annual radon concentrations within a period equal to the age, A, of the “trap”) is connected with contemporary indoor radon concentration [Yarmoshenko et al., 2005].

Exposure, dose and risk estimation

Health implications of long term radon exposure

As described above, radon concentrations were measured in many dwellings in the investigated areas. In many cases the concentrations were well above what are considered as unacceptably high levels from a health perspective for human exposure. For example in many countries there are ranges of radon action or reference levels above which it is recommended that remedial action should take place to reduce high radon exposures. These actions or reference levels differ from country to country but are generally in the range 200 to 600 $Bq \cdot m^{-3}$ [ICRP65, 1994]. It is therefore necessary to estimate the lung cancer risk of the population in these communities, where indoor radon levels exceeded the action or reference levels.

Radon is one of a very small number of substances which have been confirmed to be human carcinogens based on human studies. It is considered Group 1 and Group A carcinogen as per the classifications used by the World Health Organization (WHO/IARC) and US Environmental Protection Agency [US EPA, 1987], respectively. The principal adverse health effect arising from the inhalation of radon and mainly its decay products is lung cancer. From a health perspective the most significant are radon (^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po) and thoron (^{216}Po, ^{212}Pb, ^{212}Bi, ^{212}Po) progeny. These radionuclides, unlike radon, shortly after their formation attach themselves to aerosol particles, while a small fraction of them remain in unattached form, depending on aerosol size and concentration and ventilation rate [Nazaroff et al., 1988]. For the risk assessment of general population radon exposure (as distinct from occupational exposure) two different approaches can be used to estimate the lung cancer risk arising from exposure to radon decay products :

One is the dosimetric approach, in which the radiation doses to lung tissues is estimated and the associated risk is evaluated using currently accepted dose/risk factors for ionizing radiations. The other is the residential epidemiology approach, which is based on the case-control studies of the general population to derive risk factors. In present study the risk to the exposed populations in the investigated areas of Serbia were estimated and compared using both approaches.

The dosimetric approach

In dosimetric approach, the dose to the lung is estimated using models of lung deposition of radon progeny and dosimetry. A number of such models exist and while they are similar in many regards they differ in a number of key assumptions such as: the identification and location of the sensitive cells in lung and the weighting factor for alpha radiation. In addition, at individual person level physiological factors such as lung morphometry, breathing rate, smoking habits, lung clearance rate, solubility of radon progeny in lung etc. need to be taken into account. It was decided to estimate the doses to the general population, which includes both smokers and non-smokers, with the use of the value $65 \cdot 10^{-3}$ $mSv \cdot Bq^{-1} \cdot m^{-3} \cdot y^{-1}$ for the dose/exposure conversion factor for estimating the doses to the general population, which includes both smokers and non-smokers. It should be noted that the dose referred to is the “effective dose”. This value is derived from the work of Marsh, Birchall and Davis for equilibrium factor 0.4 and occupancy factor 0.8 [Marsh et al., 2005]. This value correspond to conversion factor $23 \cdot 10^{-6}$ $mSv \cdot Bq^{-1} \cdot m^{-3} \cdot h^{-1}$ from exposure on radon equivalent equilibrium concentration to effective dose. This value considerably exceeds the value $15 \cdot 10^{-6}$ $mSv \cdot Bq^{-1} \cdot m^{-3} \cdot h^{-1}$ estimated for dosimetric approach in UNSCEAR 2000 Report [UNSCEAR, 2000] and the value $9 \cdot 10^{-6}$ $mSv \cdot Bq^{-1} \cdot m^{-3} \cdot h^{-1}$ recommended by the same report.
For estimating the dose and risk to the general population in the investigated areas the fatal cancer risk coefficient $5 \cdot 10^{-5}$ mSv^{-1} [ICRP21, 1991] is used for estimating the lifetime risk considering 70 years of lifetime. Estimated life time risk for radon induced lung cancer in 3 rural communities in Serbia: Kalna, Gornja Stubla, Niška Banja are estimated and presented in the Table 7.1.
It is fully realized that a number of assumptions are made to derive these estimations. However, as they are not unreasonable these estimates may be taken as reasonable values at least for the purpose of comparing

the radon risk between these three areas. It cannot be overemphasized that such estimates are not meant to apply at the level of an individual person but may be useful to decision makers and health authorities to develop policies and strategies for reducing future radon exposures and associated risks in these areas keeping in view the recommendations by ICRP and IAEA to reduce radon exposure in the houses.

The epidemiological approach

In past decades, epidemiological studies on the health effects of radon exposure were made for underground miner cohorts (mainly on uranium and other hard rock miners). The results of these cohort studies have been extended to use for the estimation of risks to the general population exposed in their homes. As an example of such estimations the ICRP Publication 65 can be mentioned [ICRP23, 1993]. In this Publication the nominal mortality coefficient $3 \cdot 10^{-4}$ WLM^{-1} was recommended for radon progeny exposure both in working places and dwellings. It was supposed that this coefficient valid both to males and females in spite of the fact that only uranium miners data were analyzed.

In recent years, however, a number of major epidemiological case-control studies on the public (residential radon studies) have taken place, which gives more direct epidemiological information on the risk to the public. The detailed results of a collaborative analysis on individual data of 7148 persons with lung cancer and 14208 persons without lung cancer from 13 epidemiological studies in Europe has been published recently [Darby et al., 2006]. This is the largest and most comprehensive study of this type published in the literature to date. The excess relative risk of lung cancer per 100 $Bq \cdot m^{-3}$ was estimated to be 0.08 (95% CI 0.03-0.16) for raw data and 0.16 (95% CI 0.05-0.31) after adjusting confounding factors. The excess relative risk was not found to vary with age, sex or smoking habits. Obviously, the absolute risk of lung cancer for non-smokers is much lower than for smokers but the excess relative risk was essentially the same for both groups.

For assessment of radon induced lung cancer the official WHO statistic data presented on site www.who.int on total and lung cancer death rate in Serbia were used. According to these data the absolute lifetime risk to die from lung cancer in Serbia is about 5% for males and 0.7% for females. For assessment of lung cancer rate for non smokers the recommendations of ICRP Publication 50 [ICRP17, 1987] were used. According to these recommendations the lifetime lung cancer risk for non smokers at zero radon concentration was estimated at 0.0053 for males and 0.0037 for females. These estimations are in the good agreement with the risk estimation 0.0041 for the combine population made by Darby at al. [2006].

For the calculations of baseline lung cancer for smokers the 40% smoking prevalence for males and 30 % for females was assumed. According to the WHO data on total lung cancer death rate for the population of Serbia and assessments of the lung cancer rate for non smokers estimated the absolute lifetime lung cancer risk is circa 11% for smoking males and 3.5% for smoking females at zero or, more precisely, at the average national radon concentration.

Using the excess relative risk factor of 0.16 (given above) we can estimate the risk to smokers and non-smokers at 100 $Bq \cdot m^{-3}$ (for example) or any other radon concentrations in interest. It should be noted that such simplified risk assessment approach when the total lung cancer risk calculated by multiplying baseline risk, risk coefficient and radon concentration is valid only for low and medium radon concentrations. For high radon concentrations, especially for smokers, the effect of risks competition should be taken into account.

The classical risk assessment with the use of Lubin [Lubin et al., 1994] and BEIR VI WL models [BEIRVI, 1999] was also conducted. On the basis of these calculations, assessments of the fatal lung cancer risk in the investigated areas: Kalna, Gornja Stubla, Niška Banja are presented in the Table 7.1.

While the risks estimates using the dosimetric and epidemiological approaches are not in agreement with each other, they both indicate that the elevated radon in these communities may represent a significant health risk. It must also be stated and it cannot be overemphasized enough that all the above estimates (both using the dosimetric and epidemiological approaches) are based on many simplifying assumptions. Disregarding anecdotal non-scientific comment on lung cancer incidence in these communities, the actual values of lung cancer incidence in these communities as a function of age, sex, radon exposure, smoking habits etc. are not presently known. Nevertheless the risk estimates using either approach suggest that the

high radon levels in these three communities (and in particular in Niška Banja) may represent a significant public health risk. From a positive perspective it is worth noting that building construction regulations to control high indoor radon levels now exist in a number of European countries. For new buildings, it is possible to construct dwellings with low indoor radon levels even in areas where the rocks and soils contain elevated radon levels.

Candidate's contributions to the publications

Candidate 's contribution I : Quantification of participation in published papers

The candidate :

- *Formulated the ideas* of all eleven published papers presented as the main body of research work in her thesis,
- *Personally initiated, determined and actively participated in all and each field site measurements* and *sampling*
- *Personally participated actively in analysis of results presented in most of the published papers*
- *Personally stimulated the international scientific collaborations* in this work with scientific g*roups from many countries (Belgium, Greece, Ireland, Japan, Italy, Norway, Russia, Poland, Slovenia, India etc. The range of co-a*uthors of these paper from many countr*ies shows the fruit of this international collaboration*
- *Improved public perception in environmental radioactivity through the scientific meetings she organized, conferences for* journalists, interviews, and personal contacts at field sites .

The following table is an attempt to quantify the contribution of the candidate to the papers presented in this thesis. It is rather difficult to divide the various contributions into separate categories as there is always overlap between them. Apart from the details given in this table the candidate can confidently state that her contribution to the papers was in most cases at least 50%. The candidate is confident that her co-authors would agree with this overall assessment.

Table EA1. Summary of the candidate's contribution regarding published papers

Number of category	Category of contribution (%)	Published papers										
		I	II	III	IV	V	VI	VII	VIII	IX	X	XI
(1)	Idea, aim of reserach	25	15	25	25	25	20	25	25	25	35	35
(2)	Field measurements and sampling	25	25	25	25	25	10	15	15	15	10	10
(3)	Laboratory work	5	5	10	5	5	10	10	10	15	5	5
(4)	Analysis of results	10	10	10	10	15	25	15	15	20	10	15
4	Contribution (%)	**65**	**55**	**70**	**65**	**70**	**65**	**65**	**65**	**75**	**60**	**65**

Legend: Categories : (1)-(4)

(1) Idea and aim of research up to 35 %,
(2) Field measurements and sampling up to 25 %,
(3) Laboratory work (etching, track counting, calibrations etc) up to 15 %,
(4) Analysis of results up to 25 %,

Candidate's contribution II: Personal vision of research goals

The candidate contributed as follows:

- To *systematic* assessment of exposure of the general population *to all components* of ionizing radiation (indoor radon, thoron and external penetrating radiation), on the basis of different types of measurements, so that an assessment is made (at least for two rural communities investigated) of the *integrated environmental exposures received by the population in these investigated rural communities*, and thus the baseline study of the investigated communities is obtained,

- To the verification, *mapping and estimation of the geogenic origin* of the elevated indoor radon and thoron areas,

- *To the identification and comparison of levels of Radon/Thoron and gamma radiation* in the investigated area specifically low and high background areas,

- *To the initiation of comparative studies of the environmental radiation baseline levels* in non-affected and affected areas in the investigated areas in the years 1995 and 1999,

- *To the initiation and stimulation of scientific collaboration* with both national and international colleagues in the area of high natural radiation exposure assessment

Candidate's contribution III: Implementation of the above mentioned categories contributed to the following:

- preparing successful research project proposals and actively participating in three such Projects supported by the Ministry of Science and Technological Development of Republic Serbia (Project 1965: 2002-2006, Project 141019B: 2007-2010 and Project 23040 2008- 2009)

- the scientific investigation of the part of the INTAILRISK and INDUWASTE projects within European Union Frame Work Programme 6 (EU FWP 6, 2004-2007) on Natural Radiation Environment (NRE)

- the establishment of bilateral scientific state projects with Slovenia(2004-2006), and Greece (2005-2007),

- the establishment of an agreement on scientific cooperation with Japan (2002-2006), Poland (2005-2007), Russia (2006-2008)

- the establishment of exceptional fruitful scientific cooperation research groups in Ireland, Norway, Belgium, Japan, Greece, Italy, Albania, India, Austria, Russia, Poland.

- the organization of two scientific Electro Chemical Etching Workshops (ECE I Workshop in 2003 at Belgrade and ECE II Workshop in 2005 at Niska Banja). These workshops were attended by many research participants from outside Serbia and resulted in increased scientific collaboration with such participants.

Conclusions

The indoor radon levels in Serbia and some part of Balkan region for dwellings in different rural communities follow a log-normal distribution (GSD=1.8–3.2). The recorded radon levels strongly depend on the type of underlying rock and the average radon levels were found to vary from 45 $Bq \cdot m^{-3}$ for limestone (Montenegro) to 1560 $Bq \cdot m^{-3}$ for travertine (Niška Banja) [Žunić et al., 2000; Žunić et al., 2002; Žunić et al., 2005; Žunić et al., 2007]. Highest measured average indoor radon concentrations were about 1200 $Bq \cdot m^{-3}$ in Niška Banja, 500 $Bq \cdot m^{-3}$ in Gornja Stubla and 200 $Bq \cdot m^{-3}$ in Kalna. As estimated by parameters of log-normal distribution, the percentage of houses with indoor radon concentration above 600 $Bq \cdot m^{-3}$ were 46% in Niška Banja, 23% in Gornja Stubla and 1.4% in Kalna.

In the areas investigated it was found there is the tendency of increasing indoor radon concentrations related to the age of the dwellings, but retrospective radon measurements give the different values from the expected value of the decade increasing coefficient. Volume trap technique gave higher value of the decade increasing coefficient than surface trap technique [Žunić et al., 1999; Paridaens et al., 2000; Yarmoshenko et al., 2005; Žunić et al., 2007; Žunić et al., 2001; Paridaens et al., 2002]. It may be due to the significant influence of aerosol concentration on the assessment of retrospective value of radon concentration by surface trap technique.

The seasonal variations relating summer and winter indoor radon concentrations in the investigated areas can be described by a linear regression model ($C_{sum} = k \cdot C_{win} + A$) with the typical value of the slope factor between the cold/warm season concentration k~0.2 – 0.6 and the intercept parameter A = 3 - 40 $Bq \cdot m^{-3}$ [Žunić et al., 2002; Žunić et al., 2005, Žunić et al., 2007]. It is proved (Niška Banja) that there is negative dependency between the number of smokers in the dwelling and the average radon concentration and this fact should be taken into account during the epidemiological studies [Yarmoshenko et al., 2005, Žunić et al., 2007]. In some of the investigated areas (Gornja Stubla) the indoor thoron levels are significant and should be taken into account during both radon measurements and radiation dose and risk assessment [Žunić et al., 2000; Žunić et al.; 2004, Žunić et al., 2005; Žunić et al., 2006].

1 Introduction – Basic information on radon and products of its disintegration in the environment

1.1 Radiation exposure to humans

During their lifetime human beings are exposed to a variety of radiations both ionizing and non-ionizing, and both from natural and artificial sources. By geochemical processes naturally occurring radionuclides as an integral part of the geo-sphere are dispersed or concentrated in different parts of the environment, depending on their geochemical properties. The level of natural background radiation is determined by the type and concentration of radionuclides, by characteristic geochemical processes in a given medium, and in the case of cosmic radiation by the altitude and geomagnetic latitude. [Dangic, 1992]

Increased concentration of natural radionuclides in the environment results in " anomalous zones" where the level of natural background radiation is increased in respect to the surroundings, in extreme cases creating radiation risks for the resident population. Therefore the natural background radiation to humans may be caused by the presence of radionuclides in the litho-, hydro-, and atmosphere, i.e. due to the earth's crust (radionuclides in soil), by cosmic radiation, or by natural radionuclides (mainly Potassium-40) in food. Thus, ionizing natural radiation of various origins and properties are present in the environment.

In the reports of the United Nations Scientific Committee on Exposure to Atomic Radiation (UNSCEAR reports) it has been estimated that excluding doses from radiotherapy and nuclear accidents, the global average annual effective dose to a member of the public is about 3.0 mSv, The various contributions to this dose are illustrated in the pie chart from the UNSCEAR 2000 reports (Figure 1.1).

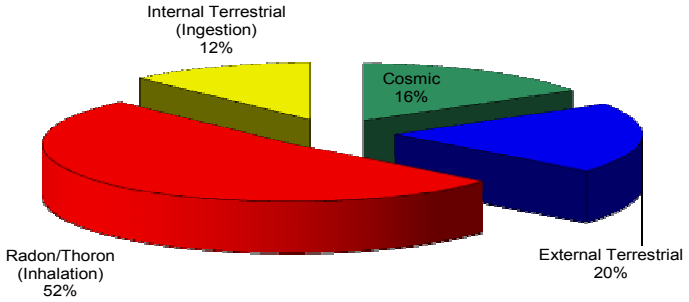

Figure 1.1 *Average exposures from natural radiation sources [UNSCEAR 2000]*

The largest contribution to the annual average dose at about 85% of the total from all sources is due to natural radiation sources such as: cosmic radiation (16%), primordial radionuclides in diet (12%), external penetrating radiation from our surroundings (20%) and from radon (52%). The rest of 15%

belongs to artificial sources: medical (14 %), nuclear discharges (< 0.1 %) ,industrial and consumer products (< 0.1 %), occupational (0.2 %) and fallout (0.2 %),. It should also be noted that, while other natural radiation doses, such as from cosmic radiation and local geology, will exhibit variations over the planet, that the component with the greatest variability and range of values is due to radon (or more correctly due to the alpha particle irradiation of lung tissue due to inhaled short-lived radon progeny). This important exposure to radon for most people is dominated by the exposure they receive in indoor environments such as their homes, schools, and workplaces. Knowledge of the behavior of radon progenies in the air is important because their contribution to the radiation health risk is considerably higher than that of their parent radon gas [NCRP, 1988].

Radon is one of the few agents for which it has been proved on the basis of human epidemiological studies that it causes lung cancer to the human population. In 1996 it has been classified as such by the World Health Organization (WHO) as being a Class I A carcinogenic agent [IARC, 1998; WHO, 1996]

In the total annual effective dose received by the population the contribution of radon is particularly significant in the anomalous zones where the level of radon and other natural components of radiation are elevated [WHO, 1996]. For this reason when the quality of indoor air inhaled is considered, radon is the fourth agent, beside asbestos, tobacco smoke, and fibrous glass whose measurement and control is required in a number of circumstances in most countries of European Union [WHO, 1996]

The problem of the effects of natural ionizing radiation upon the general population is quite complex and belongs to the scientific area of interaction of high-energy radiation with living tissue, i.e. the biological effects of radiation. If ionization occurs in living cells it may damage them by inhibiting normal cell function, preventing reproduction or even killing them. In the case of humans, this can lead to harm to organs in the form of cancer or even death. The biological effect of radiation stems mainly both from its direct effect on the molecule of deoxyribonucleic acid (DNA) and indirectly due to free radical production as a result of its radiolysis of water molecules in tissue (Holian and Garrison, 1967) Since molecules of DNA are large, they make targets of a high cross section for interaction with radiation [Ward, 1981;Brunjolfsson and Kaprielian-Matematicki, 1977].

The degree of radiosensitivity grossly varies within individual specie, if individual sensitivity is concerned. The problem of radio sensitivity occupies a central position in radiobiology which by its scientific philosophy covers the targeted research value of this work. The first phase is investigation of population exposure to radon and the accompanying components of ionizing radiation from nature.

Data on radiation sources and their contribution to the total dose that an inhabitant of a country continuously receives is usually achieved through a national survey of indoor radon. Many countries in Europe have carried out extensive geographically and/or population based surveys of indoor radon [UNSCEAR, 2000, Dubois 2005]. Areas of high indoor radon levels have been identified in all of these countries [Ghiassi-Nejad et al., 2004].

Since 1980 in the area of radiation protection and general health of population, not only in the countries of Western and Central Europe, but also in North America, and Japan national programs for radon surveys were carried out concerning the exposure to the natural radioactive gas radon and its radioactive progenies and thoron [Simopoulos and Scyvier, 2000; Katase and Michikuni 1998; European Commission 1997; Campus-Venuti, 1994; Janssens et al, 1992].

Within the scope of these investigations, in addition to national programs, there are numerous research projects supported and financed by the European Commission (1997). These programs include epidemiological studies, lung dosimetry, preventive measures and technologies for reducing radon concentrations, and development of retrospective methods of measurement of the exposure to radon in indoor air spaces. Some of the programs make estimates of radiation risk (the concept of radiation risk is

the probability that the irradiated subject suffers a consequence due to a random radiation effect).

Through national programs for radon in the majority of European countries the concentrations of indoor radon have been determined. The recommended concentrations in the air for new buildings are 200 $Bq \cdot m^{-3}$ *and 400-600* $Bq \cdot m^{-3}$ *in old buildings [ICRP 1993; ICRP 1984]. The Europpean Commission recommended mitigation in buildings with more than 400* $Bq \cdot m^{-3}$ *[EC 1990]*

In the investigated area there are uranium deposits, radioactive anomalies of uranium and thorium, and uranium ore sites (Figure 1.2) [Jankovic, 1990; Antonovic 1978; Jankovic 1965]. The reasons for the existence of high indoor radon levels are many and complex. They include geological factors, building characteristics, and usage patterns, etc. To the present time in the case of Serbia there have been very few investigations to identify areas with above normal natural radiation levels and in particular those with the potential to have high indoor radon levels. Based on a geochemical approach, this work presents the theoretical basis, strategy, methodology, and results of an investigation of population exposure to indoor radon in three investigated high radon rural areas.

The results are presented of experimental (field) work carried out in two uranium ore rich and one radium water rich rural communities in Serbia, i.e., Kalna in Eastern Serbia, former uranium mine, Gornja Stubla in Kosovo and Niška Banja in South Serbia, respectively [Zunic et al 2009; Zunic et al., 2007a and Zunic et al. 2007 b; Zunic et al., 2006; Yarmoshenko et al 2005; Paridaens et al 2002; Paridaens et al 2001, Zunic et al 2001; Paridaens et al 2001; Zunic et al.2000; Zunic et al. 1999; Jakupi et al., 1997].

Geological studies in this region have clearly identified sites which have above normal levels of uraniferous material but little or no investigations on the exposure of humans to natural radiations in these areas have heretofore taken place. The work described here addresses this knowledge deficit and as will be shown below does indeed identify and assess the dose/risk impact in a number of high natural radiation areas mentioned above.

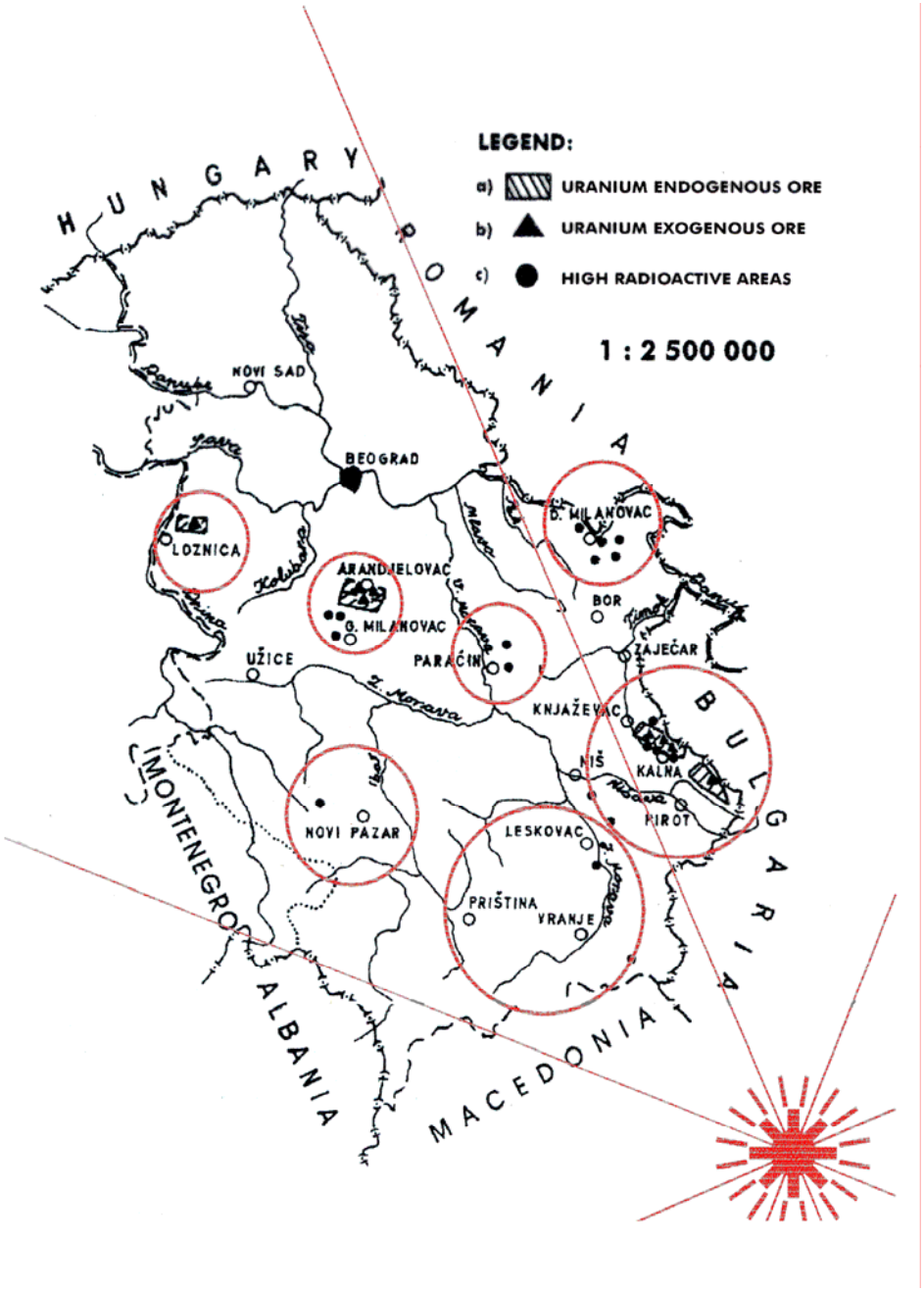

Figure 1.2 *Map of uranium ore fields, deposits and occurrence [Jankovic, 1990; Antonovic 1978; Jankovic 1965]*

1.2 Components of exposure

The exposure to humans can vary greatly from person to person depending on their proximity to radioactive sources (both natural and man-made), altitude and their living and working environment. The sources of radiation exposure to humans worldwide, arising from both man-made and natural radiation, are outlined below and highlight the large contribution from radon gas.

1.2.1 Natural radiation

Natural Radiation is the most significant contributor to human exposure. This can be divided into a number of different categories, which already are outlined in Figure 1.1.
Cosmic radiation exposure consists of cosmic rays and cosmogenic nuclides. The majority of cosmic rays that reach the earth originate within the earth's own galaxy and the major

contributor to this exposure, at ground level, are muons. Cosmic rays also interact with the atmosphere to produce cosmogenic nuclides that include ^{3}H, ^{14}C and ^{22}Na but contribute very little to the overall exposure. However, aircraft passengers and, in particular, aircrew are exposed too much higher levels of cosmic radiation than the general population. These dose rates vary significantly with altitude, latitude, flight path and solar activity (Kelly et al., 1999; Tommasino, 1999)

Terrestrial radiation is present in all media, including the human body. The external terrestrial exposure is mainly gamma radiation arising from the ^{238}U and ^{232}Th decay series and from ^{40}K present in the environment [UNSCEAR, 1993]. These radionuclides also give rise to internal exposures in the human body via inhalation and ingestion of these and their progenies. Ingestion occurs due to their presence in foodstuffs and drinking water and accounts for the internal terrestrial exposure in Figure 1.1.

The largest contributor to natural radiation exposure, and overall radiation exposure, is due to the inhalation, and subsequent deposition, of the short-lived ^{222}Rn and ^{220}Rn progeny in the lung. Until relatively recently exposure from radon was not considered significant enough to have a detrimental effect on human health in the general population but this perception changed with the discovery that some residences had indoor radon levels comparable to those in underground mines [ICRP, 1993].

1.2.2 Anthropogenic radiation

Human-made radiation makes up 15% of the overall exposure to humans worldwide (UNSCEAR, 2000), and consists of:

- *Medical radiation procedures*
- *Atmospheric and underground nuclear weapons testing,*
- *Nuclear power production,*
- *Nuclear weapons fabrication,*
- *Radioisotope production and use.*
- *Nuclear accidents.*

Medical radiation procedures are the largest man-made source of radiation exposure to the world's population, accounting for 14.2% of the overall exposure to humans, and are likely to increase worldwide as developing countries improve their medical services [UNSCEAR, 1993)

The exposure due to nuclear weapons testing accounts for less than 0.2% of the total and has been decreasing since the early 1960s, the end of the most intensive weapons testing period. Nuclear power production, weapons fabrication accidents and radioisotope production and use contribute the remaining 0.1% of the man-made exposure.

1.3 Radon gas and its properties

The element Rn is an ubiquitous gas. Much has been written on the subject [Proctor 1995; Stannard 1988,] and a brief history is given in Table 1 outlining its discovery, as well at its path from being considered only as an occupational hazard to underground miners to being a major source of exposure to the general population.

Radon is a colorless, noble gas consisting of three isotopes: ^{222}Rn, ^{220}Rn and ^{219}Rn arising from the natural decay schemes of Uranium-238 (^{238}U), Thorium-232 (^{232}Th) and Uranium-235(^{235}U), respectively. By radioactive disintegration of uranium (Figure 1.3) and thorium (Figure 1.4) different isotopes of radon are formed, the most stable, most widespread and the longest-lived isotope is ^{222}Rn, (later in this text this isotope is referred to as "radon"), which arises by the disintegration of ^{238}U, i.e. immediately upon disintegration of ^{226}Ra, having a half life of approximately four days (3.824 days), and can significantly influence the radioactivity of air, water, or soil. The abundance of another radon isotope ^{220} Rn usually called thoron (a progeny of thorium) is four times higher, but since the half life is approximately one minute (54 s), its presence in the air is of minor influence. The third radon isotope , actinon or ^{219}Rn, has a half life of 4s and its contents in the soil is small and the influence of this last isotope of radon on radioactivity of the air can be neglected. Two other isotopes thoron (^{220}Rn) and actinon (^{219}Rn) have very short half-lives, and contribute very little to human exposure when compared to radon and are not included in this study.

Rn-222 is the only isotope in the uranium decay chain that is a gas. Furthermore, it is an inert gas with a half-life sufficiently long enough to allow it to travel, via diffusion and advection, through materials. Disintegration of radon-222 (^{222}Rn) results in short lived alpha and beta progenies polonium-218 (^{218}Po), polonium-214 (^{214}Po), and bismuth-214 (^{214}Bi) and long lived lead-210 (^{210}Pb) and polonium-210 (^{210}Po) and bismuth -210 (^{210}Bi) between them.

Most of the radon formed in the soil can reach the outdoor atmosphere from a ground depth of up to two meters. This depends upon a number of factors that include geology, climate, moisture, and dynamics of release and recoil and soil characteristic, mainly permeability. Typically, concentrations of radon in soil gas range from 10 000 $Bq \cdot m^{-3}$ to 50 000 $Bq \cdot m^{-3}$ but once entering the outdoor atmosphere, the radon is dissipated over such a large volume, thus the average outdoor concentration ranges from 1 $Bq \cdot m^{-3}$ to 100 $Bq \cdot m^{-3}$ [UNSCEAR, 2000]. In general, indoor concentrations substantially exceed those outdoors, ranging from 6 $Bq \cdot m^{-3}$ to as high as 85 000 $Bq \cdot m^{-3}$, making indoor air the largest single component of natural radiation exposure [Swedjemark et al., 1993].

Although radon in indoor air arises from the radium content in the earth's crustal materials it may enter dwellings through a number of different pathways.

Table 1.1 *A brief history of radon gas and epidemiology*

16th Century	*An unusually high occurrence of what we now call lung cancer was noticed amongst miners in the regions of Schneeberg and Joachimstal (Jachymov) in Bohemia (Czech Republic).*
17th Century	*There was an increase in the frequency of this disease, now known as Schneeberger Lungenkrankheit, as the mining in these regions intensified.*
1879	*Haerting and Hesse identified the Lungenkrankheit as lung cancer.*
1898	*Marie and Pierre Curie extract ^{226}Ra, radium, and ^{210}Po from Jachymov ores.*
1900	*Radon identified by F.E. Dorn as a radioactive noble gas produced by the decay of radium, it was initially called niton but its name was changed to radon in 1923.*
1901	*First radon measurements carried out in Schneeberg and Jachymov by Elster and Geitel.*
1911	*The link between high radon levels and lung cancer was first assumed by H.E. Müller*
1940	*On the basis of radon measurements in the mines in Schneeberger and biological studies made on miners who had died from lung cancer in this region it was concluded that radon was a possible cause of the high incidence of lung cancer amongst the miners in this region.*
1951	*After numerous studies failed to explain the high incidence of lung cancer due to radon gas alone W.F. Bale proposed radon progeny may be the principal cause.*
1957	*The 'Working Level' concept was introduced in order to better quantify the dose received to miners from radon progeny.*
1956	*The first residential radon gas measurements were made in Sweden.*
1970 to present	*Larger surveys on indoor radon made in several countries reveal indoor radon levels in some dwellings comparable to those of underground mines. A number of epidemiological studies were and are being carried out in order to find the excess risk to humans.*
1988	*Radon classified a human carcinogen by IARC [IARC 1988]*
2000 to present	*Recent residential radon epidemiological studies have yielded reliable estimates of the radon risk to the public in their own homes, outlining its discovery, as well at its path from being considered as an occupational hazard to underground miners to a major source of exposure to the general population. Thus, confirming the need for studies in residential radon epidemiology.*

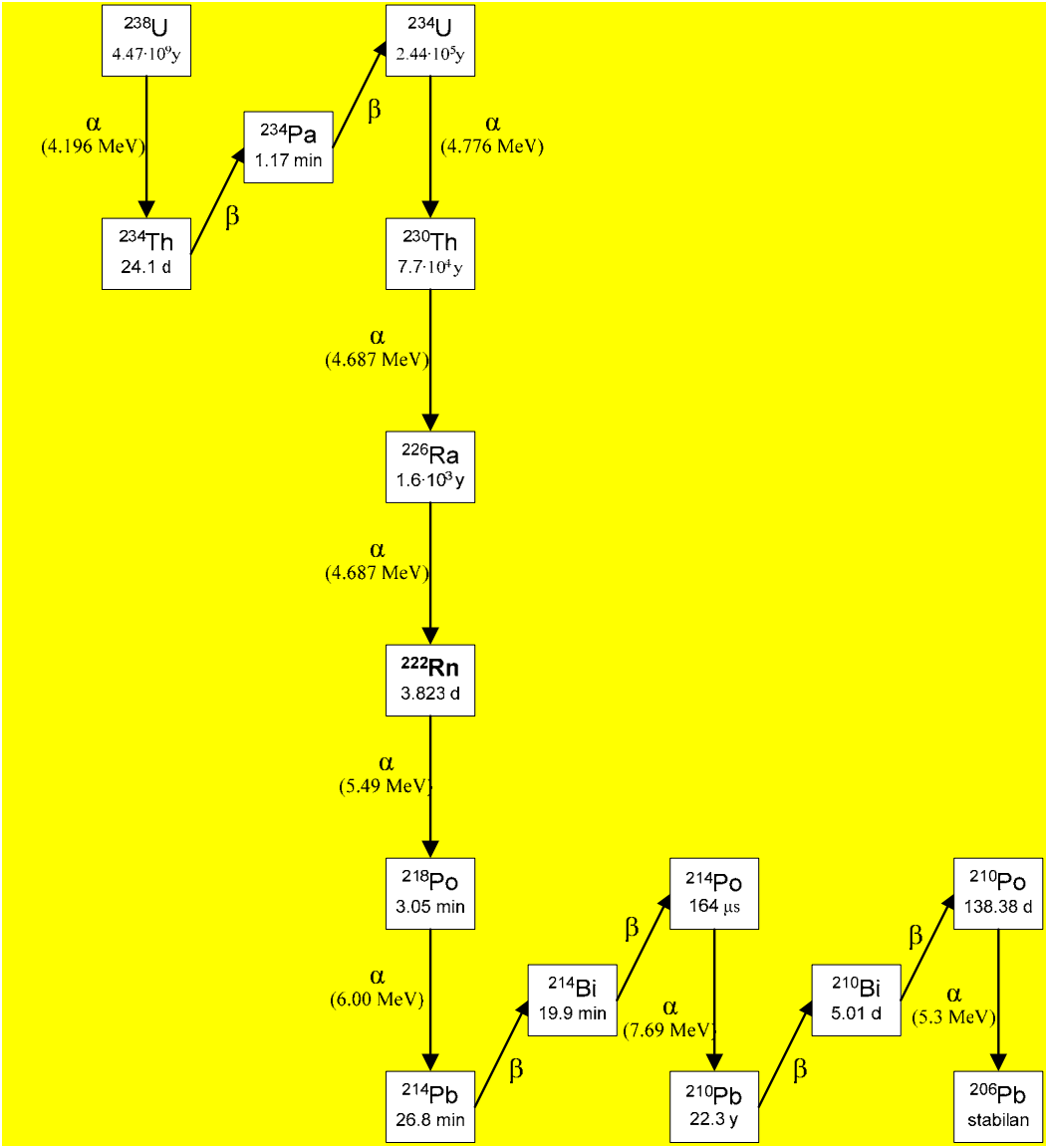

Figure 1.3 *The ^{238}U decay chain displaying the major radiations. Arrows pointing downward denote decay by α-particle emission. Arrows pointing up- right indicate decay by β-particle* emission.

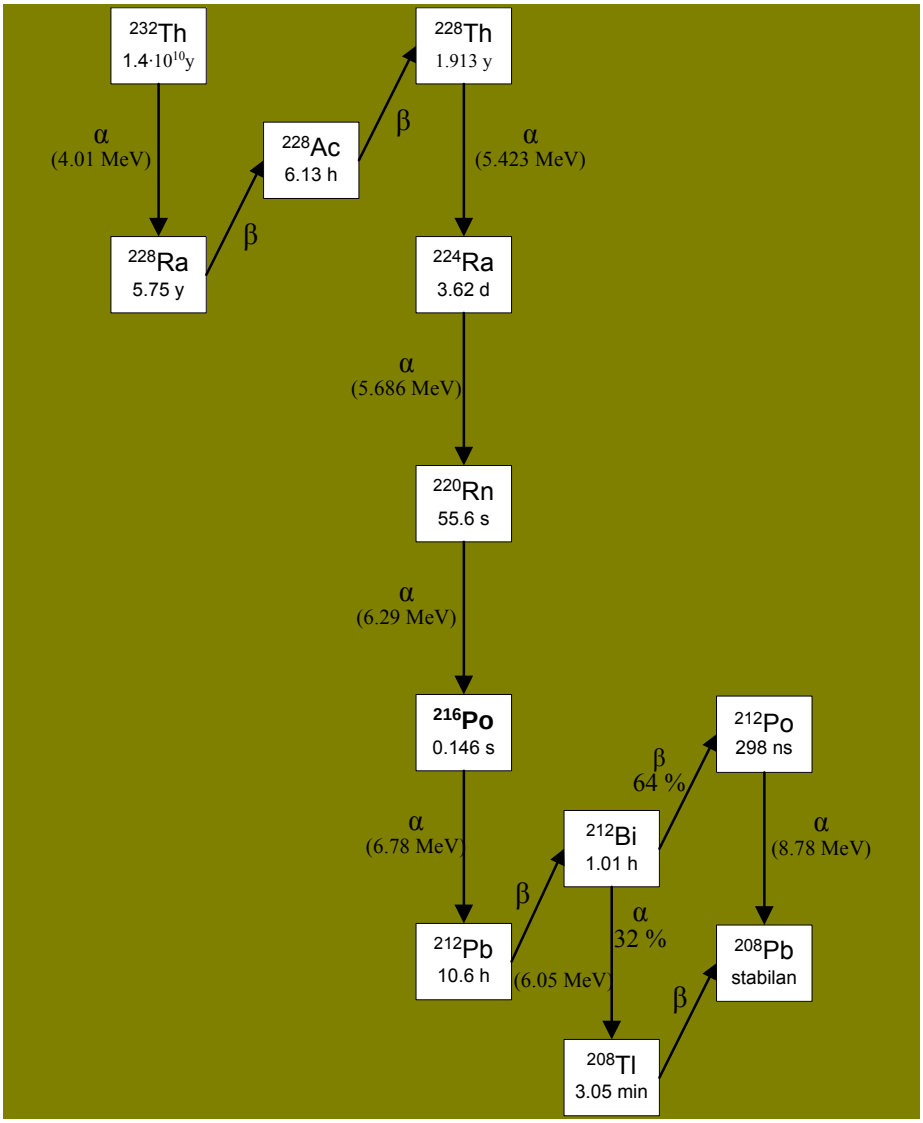

Figure 1.4. *The ^{232}Th decay chain displaying the major radiations. Arrows pointing downward denote decay by α-particle emission. Arrows pointing up- right indicate decay by β-particle* emission.

2 Radon and products of its disintegration

2.1 Sources of radon in indoor air and its propagation

Radon is present in indoor air spaces owing to the difference that usually exists between the external and internal pressures, i.e., usual small pressure differential between the indoor air and the subjacent soil gas [de Meijer et al., 1992]. This pressure difference causes it to penetrate through small holes in the foundations of the building. Radon in indoor air arises from emanation from the soil and building materials and, to a lesser extent, from household water if it originates from local source, i.e., subterranean water containing high concentration of radon and from gas. Radon also penetrates from the outdoor air which may be of importance if the concentration of radon in the atmosphere is high. Therefore, radon concentration in closed spaces is a function of several parameters, and the relevant importance of above mentioned pathways varies with the dwelling. The most significant parameters are the intensity of ventilation, total intensity of radon sources, volume of the closed space, radon concentration outside the building, meteorological conditions, etc.

As the soil beneath the house, building material and potable water supply and cooking gas can be major sources of radon in houses, a brief description of various radon sources is given. Emanation from the soil is considered as being the dominant source for ground floor dwellings whereas the dominant sources in high-rise dwellings are building materials and outdoor air [UNSCEAR, 1993]. Figure 2.1 outlines the typical radon sources and pathways into a dwelling.

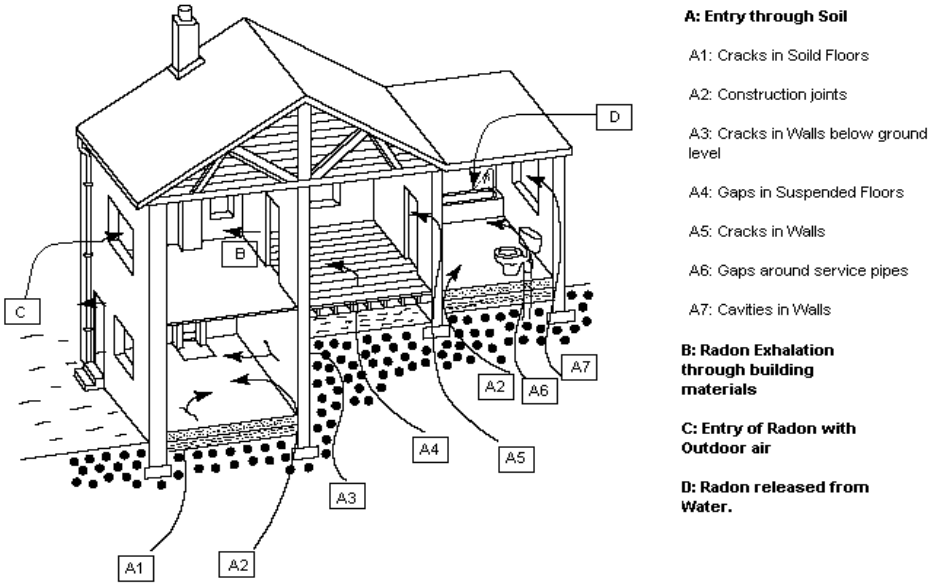

Figure 2.1 *Methods of radon entry into a typical dwelling (modified from RPII, 2004) and typical paths of radon propagation in closed spaces [de Meijer et al., 1992; Nero, 1989; Nero, 1988]*

2.1.1 Radon in soils

Soil beneath the house is considered as a major and most important source of radon inside the house, in general. A significant mechanism of radon entry to closed spaces is through cracks and holes in the foundation of a building owing to a lower pressure inside closed space compared to the outer atmosphere. Radon concentration in the soil is determined by its content of radium and radon in the rocks and foundation of the house humidity and permeability of the soil [Nero, 1989, Castren et al., 1985; UNSCEAR, 1977].

Soil moisture plays an important role in the emanation of radon for several reasons. Soil moisture in the form of a thin film of water surrounding soil grains, directly affects radon emanation by capturing the radon which escaped from the solid matrix due to its kinetic recoil energy after the decay of the Ra atom. The emanation of radon from the air filled pores of soil is higher under moisture than under dry conditions. The mechanism of radon entry in the house is based on the pressure difference, which facilitates the migration of radon from soil to the house foundation. Thus, the ingress of radon from the soil is dependent upon the radon potential of the soil and the radon flow rate into the house. As radon potential depends on radium content in the ground and the permeability of the soil, the radon concentrations of the soil typically ranges from 10 000 to 50 000 $Bq \cdot m^{-3}$ [Schumann, 1993], but the permeability of the soil has a large effect on the ability of these concentrations reaching the dwelling itself.

Soil permeability has a very broad range from less than 10^{-16} m^2 for homogenous clay to 10^{-8} m^2 for clean gravel. Grounds with a high radon potential are alum shale, granites and certain volcanic rocks due to their high concentrations of radium and high permeability [Tyson et al., 1993].

Inflow into dwellings is caused by advection and diffusion. The pressure gradient between ground and soil influence the entry of radon in the house by molecular diffusion through the cracks. The effectiveness of pressure differential in pulling in radon-leaden soil gas through the foundation is critically dependent on the effective permeability of both the building foundation and the adjacent earth.

Advection is driven by the pressure differential between the building and the ground around it. This differential arises from the higher temperatures in the building (the stack effect), ventilation and the weather outside the dwelling (mainly wind and atmospheric pressure) and causes radon to be drawn in from the ground under the building. The advection can be increased significantly with the presence of cracks, openings and joints in the building materials because of the differences in pressure differentials and permeability; the advection contribution is highly variable from structure to structure, at least in temperate and cold climate.

Studies in Finnish houses have shown that the radon entry rate due to diffusion accounts for only 6.25% (wooden housing) or 5.71% (masonry housing) of the total entry rate from the subjacent earth and this contribution decreases with increased adjective flow [Arvela, 1995].

To conclude the mechanism of penetration based on the difference in pressure moves the air, and with it moves radon, from the encountering soil up to house foundations wherefrom it penetrates the house by:

1. *Molecular diffusion*
2. *Pressure gradient between the house and soil, and*
3. *Via cracks or by water when radon can be transferred at distances over 100 m.*

2.1.2 Building materials

Radon emanation from building materials was initially considered the primary source of radon in indoor air [UNSCEAR, 1977], but measurements made in U.S. houses in 1980 found that radon emanating from building materials could not account for observed indoor concentrations; leading to the present consensus that radon emanating from soil gas is the main source of the high radon levels seen in homes. However, it is still considered as a primary source of radon gas for above ground dwellings. The radon gas in indoor air arising from building materials is largely dependent upon the radium content of the building material. Building materials can be of natural origin; such as concrete, clay or sand-lime bricks, stones or natural gypsum; or can be by-products of different industries such as phosphogypsum, slag or coal-fly ash. Typical radium concentrations in building materials, in the EU, are 40 $Bq \cdot kg^{-1}$ for natural materials and 280 $Bq \cdot kg^{-1}$ for industrial by-products [EC, 1999].

Building materials with increased radium (^{226}Ra) content and a high intensity of emanation are responsible for the elevated radon flux from the walls [UNSCEAR, 1977]. As radon flux one understands the activity of radon per unit time and unit surface of the wall. If this is combined with a poor ventilation of buildings, radon concentrations could reach high values. Building material is also a source of additional gamma radiation. In equilibrium the rate of creation of radon, defined as the number of atoms ^{222}Rn created per unit time, is equal to disintegration rate of ^{226}Ra. Due to its half life only a part of the radon atoms will come out of the material where they have been created. The others will disintegrate within the material before they reach the air. The fraction of radon in indoor air arising from building materials also depends on the ability of the material itself to transport the gas. This happens either by diffusion or by flow, where the fluids within the material act as a carrier. The factors determining the transport are the permeability, porosity and density of the material, surface preparation and finish of the wall as well as the atmospheric pressure and relative humidity in the room.

The release of radon atoms from the material where they have been created is carried out by several mechanisms (Figure 2.2). Namely, if the core of ^{226}Ra (decaying radium atoms) is on the surface, by its emission of an alpha particle one atom of radon being formed will pass about 0.1mm until it is stopped, depending upon the material where the deceleration occurs. Soon after creation radon atom will pass to atmosphere.

If an atom of radon has been formed in the interior of the material, it is necessary that it finds a crack or a pore in the material wherefrom it can reach the atmosphere. It can reach the pore space only if the distance which it has to travel is not too long, giving its kinetic energy (recoil from alpha decay) and the stoping power of the material. In order that an atom of radon becomes free, it is necessary that it stopped in a hole depending on the fluid of the hole. A hole is mostly filled by water or by air Once the Rn atom has reached the pore space, it must be stopped by air or pore water, otherwise it may collide with an opposite grain surface and be implanted into the grain. Since the stopping power of water is higher, the water stops more efficiently the retreating atoms of radon than the air. From the pore space a Rn atom can migrate into the open air by diffusion or advective transport. Sometimes it occurs that radon atom crosses a hole and re-enters the material, whereby it is lost for the diffusion in the atmosphere. A diffusion through the crystal lattice, as a path of radon atoms to the cracks and holes is possible, but this process is of less significance. Diffusive migration of Rn within the grains is a very inefficient process and can be neglected.

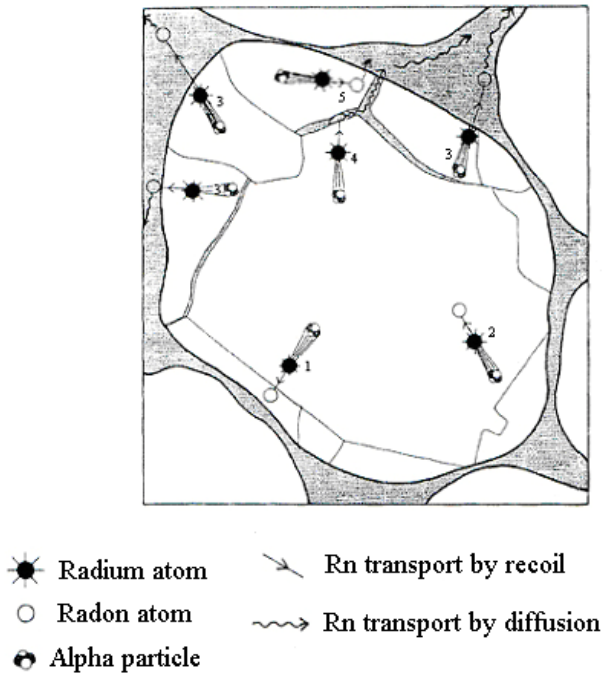

Figure 2.2 *Radon emanation mechanisms from the mineral grain (modified by Åkerblom and Mellander, 1997)*

The emanation quotient (emanation fraction, emanation power, defined as the fraction of generated Rn which leaves the material) is controlled by several quantities, such as: material grain size, distribution of ^{226}Ra in grains, microscopic structure of material, porosity of materials (portion taken by holes) and amount of water in holes (humidity). The listed parameters are not easily measured quantities. For this reason intensity of emanation and intensity of exhalation are introduced. The unity of emanation is the activity of radon (in Bq) obtained from that material, divided by the mass of that material per unit time.
The unity of exhalation is the activity of radon obtained by surface division of this material per unit time. The unity is $Bq \cdot m^{-2} \cdot s^{-1}$.

Apart from emanation power, the second most important property in respect to Rn exhalation of the building material, i.e., essential characteristic of the building material is the content of ^{226}Ra. The average content of ^{226}Ra for Europe is 33-74 $Bq \cdot kg^{-1}$ [Swedjemark and Mjones 1984; NEA/OECD, 1979].

Building materials made of industrial or mining waste could also contain an increased quantity of ^{226}Ra. Such material is red ash, created during processing of bauxite, when used for making building blocks or bricks. Average content of ^{226}Ra in this material is 280 $Bq \cdot kg^{-1}$. In practice ashes and dross from thermal power plants are used as building materials. Concentrations of ^{226}Ra in such materials could reach values of several hundreds of $Bq \cdot kg^{-1}$ [Ingersol, 1983]

The intensity of exhalation depends on wall thickness and density of the building material. The radon flux depends not only on the properties of the wall (density, chemical composition, and humidity) but also on the meteorological parameters such as atmospheric pressure, temperature, wind speed, etc.

Typically radon emanation from building materials leads to a typical excess indoor radon concentration of 10-20 $Bq \cdot m^{-3}$ but can be as high as 1000 $Bq \cdot m^{-3}$ for materials with very high radium content such as alum shale, volcanic tuffs or pozzolana [Tyson 1993].

2.1.3 Water and gas

The contribution of tap water (including showers and toilet flushes) or natural gas to radon concentration is of secondary significance. Of course, the method and extent of their use could influence radon concentration; radon concentration in water depends upon its origin, (type of water) and processing.

The concentration of radon in water may range over several orders of magnitude, generally being higher in well water, intermediate in groundwater and lowest in surface water. It is estimated that approximately 50% of radon content is freed during its use [UNSCEAR 1993].

*Radon can be readily dissolved into water and can be transported with it into dwellings where it is released into indoor air via agitation or heating. Both surface and sea waters have a very low radon concentration. Recent measurements made in seawater show radon concentrations ranging from 30 kBq·m^{-3} at sea floor level to 3 kBq·m^{-3} near the sea surface [Anders**e**n 1999].*

However, in wells drilled in rock, radon concentrations in the water may be high. It has been estimated that 1·10^{6} Bq·m^{-3} of radon in water will contribute up to 1·10^{2} Bq·m^{-3} to the indoor air concentration, and for a level above this the radon in drinking water may pose a significant additional health risk, in the long term, to users who depend on ground water supplies as their primary source of drinking water [RPII 2003].

Natural gas used for cooking and heating contains about 1·10^{3} Bq·m^{-3} of radon, which is released into indoor air on combustion. However, the contribution to the overall indoor radon concentration is thought to be negligible [Nazaroff and Nero 1988].

2.2 Radon in indoor air

In order to carry out a reliable investigation and ensure the corresponding precision and accuracy, a measurement quality assurance program has to be provided [DOE 1990; EPA, 1989; NCRP, 1988; EPA, 1987; NEA/OECD, 1985]. It comprises:

1. Quality of radon measurements (inter-calibration and inter-comparison of detectors),

2. A test of dependence of the influences of various factors on the concentration of radon and its progenies in closed spaces.

In the course of investigations concerning the quality of the breathing air it was noticed that the concentration of radon was related to the concentration of its progenies, through the equilibrium factor (F). The value of this factor is between 0.2 and 0.7 for the air in inhabited closed spaces. The usual value is F=0.5 meaning that radon concentration is double the concentration of its progenies. The value 0.4 is often used [UNSCEAR, 1993; ICRP 1993; UNSCEAR, 1988].

Variations in radon concentration depend on:

- *radon propagation (through the soil, building material, water, or air),*
- *time of day and duration of measurement,*
- *duration of detector exposure,*
- *number and locations of measuring points.*

In this chapter each of the listed dependencies will be considered.

2.2.1 Radon concentration and ventilation

Ventilation lowers radon concentration in closed spaces. Ventilation intensity is expressed as the number of complete changes of air of a closed space within one hour. Ventilation consists of infiltration, natural, and artificial ventilation. Infiltration is uncontrolled flow of air in a closed space when doors and windows are closed. Natural ventilation implies that the ventilation is done by opening windows or doors, whereas artificial ventilation is done by means of electro-mechanical devices. In the absence of artificial ventilation, ventilation intensity is one change of air per hour. Buildings having ventilation rate intensity bellow 0.5 change of air per hour are considered poorly ventilated. Ventilation intensity during summer is different from the one during winter. Calculations show that ventilation intensity is predominantly dependent upon the strength of the source of the air [Cavallo 1992; Wolliscroft 1992; Renken and Konopack 1992; DOE, 1990; NCRP, 1989; Holub, 1985; NCRP 1989].

2.2.2 Temporal variation of radon concentrations

The concentration of radon varies continuously, from short time variations to seasonal and annual variations. This concentration is affected by climatic factors, weather conditions (air pressure, humidity, speed of wind), building material of the object, heating or cooling method, and living habits. Usually radon concentration at evenings is higher than during daytime. It could be higher at early morning and early afternoon hours. In winter times radon concentrations are higher than during summer, but it is very difficult, to confirm these variations with sufficient reliability. If the measurements are carried out over long periods of time these differences become smaller; namely, very short measurements lasting several minutes or several hours could show order of magnitude differences in radon concentration whereas measurements lasting several days show differences which are smaller than that; seasonal variations of radon concentration during measurements lasting six months are different by factor 5 [Bochicchio, 1992; Hess, 1985]. Differences in radon concentration measured over a period of one year can differ by a factor of 2 [Martz, 1991]. Because of these experimental factors, it is considered that measurements of radon concentration in closed spaces in all four seasons throughout one year give the best compromise for calculating the average radon concentrations in indoor spaces.

2.2.3 Detector exposure duration

There are four main types of exposure of solid track detectors in closed spaces. Long term exposures (from several days to several months) give the best estimates for the average values. However, if the occupants are absent for long periods of time, the conditions during these measurements would not correspond to real life situations. If for some reason a quick estimate of this concentration is required, e.g. screening an area, one could apply shorter periods of measurement. Then one has to calculate carefully the factor of seasonal correction in order to determine the long term average value and this is a potential measurement uncertainty. This procedure could give reliable result only if prior to this measurement seasonal variations of radon concentration have often been measured for the same type of building, over the same area, and the corresponding results were known to lie within a narrow range. There are measuring instruments for long term radon measurements because an integrated measurement of radon progenies is difficult owing to both technical and conceptual problems. Short term measurement of radon concentration, typically 1-10 days, can be used for the purpose of quick screening and identification of low, medium, and high concentrations. Another goal of such method of measurement could be a rapid identification of closed spaces having high radon concentrations. In these cases measurement protocols are made in such a way that radon concentration is maximized for instance by sealing the house during the measurement period [EPA, 1987a]. Short term measurements cannot be used for any precision calculations of the average

values and long term measurements are necessary [White, 1994; EPA, 1992; EPA, 1987b].

An ultra short measurement, typically 10 minutes, is usually performed in diagnostic purposes in civil engineering or for making experiments in investigations of building materials. Continuous measurements are performed primarily for controlling radon concentrations in working spaces such as mines. Sometimes they are used in short time or instantaneous measurements.

2.2.4 Number and location of measuring sites

When radon concentration is determined for the purpose of investigating the exposure of population, closed spaces populated most of the day (living rooms) or where people sleep (sleeping rooms) are selected. Measuring sites inside kitchens or bathrooms are avoided because these spaces could contain high aerosol concentrations or ventilation could be intensified and the accuracy of the results could be affected. Measuring sites are located to be at average human height, i.e. at places where air is inhaled; it is also required that in the vicinity of a measuring site there is no significant air flow, a source of any heat, or an electrical appliance which might affect the measurement.

When houses or flats are examined, then closed spaces where air is the least ventilated are chosen; in houses these are usually ground floor rooms, or basements, or cellars. Measurements could be organized in such a way as to determine the paths of radon in-flow. The numbers of closed spaces and measuring sites are dependent on the size and type of a house or a flat.

Depending on the size or type of a house (i.e. whether there are one or more floors) and also on the number of rooms (number of living rooms and bedrooms), radon concentration could be measured in bedrooms in case of an one floor house or in two or more rooms in cases of two- or several floor houses.

When workplaces are concerned, such as kindergartens, schools, hotels, storehouses, banks, and other business premises, depending on the size of the building, sizes and number of rooms, type of building construction, ventilation and heating systems, the selection of rooms is made and radon concentration is determined. It is usual that a single site is covered by one detector, but sometimes several detectors are placed in one closed space. In this way a better reliability of the measurements is ensured, which has been carried out in this study in several cases of houses in rural communities.

2.3 Radon in outdoor air

In the atmosphere emitted soil gas radon becomes diluted with estimated outdoor levels being, on average, only 10 $Bq \cdot m^{-3}$. The outdoor level can change diurnally and seasonally, as it is dependent upon the time of day and the weather. Solar heating during the days leads to radon being transported more readily from ground level whereas at night time atmospheric inversion causes the radon to be trapped close to the ground. Seasonal variation is related to the effects of precipitation and changes in the prevailing winds.

Coastal regions and islands, such as Ireland, can have outdoor levels as low as 1 $Bq \cdot m^{-3}$ as the air over oceans has a very low radon concentration (~ 0.1 $Bq \cdot m^{-3}$). These levels can raise to as high as $1 \cdot 10^{2}$ $Bq \cdot m^{-3}$ for areas near substantial radon sources such as mine tailings.

The overall contribution from outdoor air to radon in indoors for a typical home is only thought to be about 20% for a ground floor dwelling but rises to 50% for high rise apartments (UNSCEAR, 1993).

Land masses are the sources of outdoor radon while oceans act as sinks. As a consequence outdoor air

radon levels are much lower (<< 1 Bq·m^{-3}) in oceanic air than over a continental land mass such as mainland Europe. Outdoor radon levels are determined mainly the soil characteristics (uranium/radium content, porosity and the consequent radon flux), local topology and meteorological conditions. In some situations such as atmospheric temperature inversions in valleys with high radon fluxes from the soil short-term outdoor radon levels as high as some hundreds of Bq·m^{-3} have been observed. High outdoor radon levels are rare but could be of local health significance in communities in areas such as former uranium mining districts where elevated radon exhalation from tailing ponds combined with meteorological and topological conditions could give rise to high outdoor radon levels of seasonal duration.

In general, however, while national data on average outdoor radon levels in Europe are quite limited it seems that they lie between 4 and 10 Bq·m^{-3} with the former value being appropriate to island nations such as Ireland and the UK and the latter more representative of the European mainland. If, as an illustration, the average outdoor value in some hypothetical mainland European country is about 10 Bq·m^{-3} where the indoor average is 80 Bq·m^{-3} then the radon risk to an individual from outdoor radon exposure might be about 6% of their risk from indoor radon.

A. I D E N T I F I C A T I O N

3 Geological origin of radon in dwellings of the investigated rural areas (Kalna, Gornja Stubla, and Niška Banja) in the Balkan region

The field locations were selected on the basis of geological-structural and geochemical prospecting data on natural radionuclides and other mineral resources found in these three rural areas. Since 1951 uranium contents of the rocky areas of Eastern, South-Eastern and South Serbia have been studied systematically by regional geological and air-radiometric investigations [Jankovic, 1965]. Several hundreds of zones of anomalously elevated radioactivity have been discovered. A total of 279 anomalous zones have been identified. Of those 146 are due to uranium, 30 due to the mixture of uranium-thorium, and 103 due to thorium [Jankovic, 1990] In these zones the natural radionuclides present to a significant degree are : carbon (^{14}C), potassium (^{40}K), thorium (^{232}Th), and uranium (^{238}U) together with their products of disintegration: radium (^{226}Ra) and radon (^{222}Rn). Carbon (^{14}C), potassium (^{40}K), thorium (^{232}Th), and uranium (^{238}U) belong to the group of soil-oriented elements.

Radium and radon follow the primary and secondary deposits of uranium, but often they form separate aureoles of dispersion owing to certain differences in their geochemical characteristics. Radium as a soluble cation may be mobile, even when uranium is bonded i.e., chemically fixed, by migration with ground water, so in waters or in soil it can be quite separated from uranium and locally concentrated. Thus, ecological processes can lead to high local concentrations of radium.

Radon is a gaseous radionuclide which is soluble in water. It can migrate easily in gaseous or liquid phase, and under favorable conditions (fault zones, soils, etc.) can reach high concentrations.

Minerals are tied to some sandy layers in sediment formations and could contain uranium in quantities from 100 to 3000 $mg \cdot kg^{-1}$ one could find autinit or other secondary minerals, sometimes pitchblende, pyrite, and marcasite (Dangic, 1987).

3.1 The rural community of Kalna

Kalna is the former uranium mining districts which have been exploited from 1948 to 1966, when the mine was closed. It is situated in the region of Stara Planina mountain at the Eastern part of Serbia, where the discovered uranium mineralization is mostly limited to Western slopes of its central part , draining towards the river Trgoviski Timok through its tributaries the Crnovrska, the Inovska and Gabrovnicka rivers. The most important ore occurrence and uranium ore deposits are ranging from 300 – 1000 meters above sea level, limited to the granite massive of Janja. The hydrothermal lentice like layers of deposits and the appearance of uranium are connected to a number of times activated zones of cleavage. To this genetic type belongs the deposit of Mezdreja. Deposits of Gabrovnica are clay like, crumbling, near-surface cleavage zones. Generally, the uranium content in the granite averages between 6 $mg \cdot kg^{-1}$ to 20 $mg \cdot kg^{-1}$.

The discovered deposits of uranium minerals cover predominantly the western slopes of central part of the mountain Stara Planina which are drained to river Trgoviski Timok by tributary rivers Crnovrška river, Inovska river, and Gabrovnička river. In the area of Stara Planina which is a significant metal-genetic

zone, there are four uranium deposits which were temporarily exploited (Jankovic S., 1965). The most significant uranium deposit in the vicinity of Kalna is concentrated at the granite massive of Janja. Hydrothermal veinal, saline deposits with appearance of uranium are tied to several times active breaking zones. To the same genetic type belong deposits of Mezdreja. Deposits of Gabrovnica are tied to clay and porous surface layers of breaking zones. Granite massive of Janja ("Janja-Inovo-Gabrovnica") is elongated along direction North-West – South-East. It is about 19 km long and about 2.5 km wide. Along the perimeter of the massive granite, magma has been contaminated which is exhibited by an increased content of biotite and rocks, while the quantities of K-feldspar and quartz are decreased. Deformations and secondary changes, noticed for the massive as a whole, are particularly exhibited along its perimeter. They appear as schistose textures, crushing of materials, and crystallization of secondary minerals. They consist of quartz, oligoclase, K-feldspar, and biotite. Subordinate constituents are sphene, apatite, zirconium, and magnetites and secondary are sericite, chlorite, epidote, calcite, limonite, and clayey substance. In the massive of Janja all transitions have been observed, from simple crushing of feldspar and quartz, then bending of mica leaves and feldspar laminae, crumbling of feldspar and filling of cracks by the crumbled feldspar, chlorination of biotite and formation of cataclastic structures, up to formation of schistose textures with sericite along schistose surfaces. Thus a rock looks like gneiss. These schistose granites are particularly well noticed in Inovo and along the southern part of the perimeter towards Balta-Berilovac.

3.2 The rural community of Gornja Stubla

This region represents the Northern branches of Skopska Crna Gora at the extreme south of Serbia. The rural area lies over Paleocene rocks, lower cretaceous flysch and granite. It borders a North-West-South-East fault zone filled with brecciate rock mass bearing secondary uranium mineral autinite. Morphologically this terrain is covered by hills with altitude from 500 up to 1000 meters above sea level. The immediate zone of Stublovača hill above Gornja Stubla community is 984 m above sea level.The soil is built up of diabasic formation (Jura), lower chalk flysch, Paleocene sediments (layers) of trachyte and trachyte tuffs. The diabase-chart formation is presented by magma and sedimentary rocks. In the Stublovaca region diabase are more abundant than clays, sands, lapor and chalk. In parts of diabase, trachytes enter deeply. The flysch fascia is composed of sand with intercalations of clays and lapors. In the structure of sandstone material formed by diabase destruction predominates. South West part of the area is built of paleogenic sediments made of mixture gray marl and clay. The hydrographic net is well developed. A large number of streams flows radially from mountain Stublovaca and gravitates toward Binacka Morava River. The springs are very frequent particularly at joints of trachyte and sedimentary rocks.

3.3 The rural community of Niška Banja

The Niška Banja vicinity belongs to the mountain range known as the Karpato-Balkan arch. It is a long structure stretching from central Europe through East Serbia, Romania and Bulgaria. The Karpato-Balkan arch of Serbia is a mountain region with certain mountain tops reaching above 2000 meters. It has a diverse geological composition and complicated tectonics, and it is well known for many mineral deposits, i.e., metals (copper, gold), non-metallic resources (building materials), coal (brown), thermal and mineral waters.

Niška Banja is located in the east part of Nis Neocene basin on the southern slopes of Koritnik (Koritnik is a part of Suva Planina Mountain and northward from main equatorial Miocene fault zone. In the geological structure of Niška Banja vicinity, rocks of several system involve Upper Devonian and Lower

Carboniferous, Permian, Upper Jurassic, Lower Cretaceous, Miocene, Quaternary (Figures 3.1 and 3.2).

In a narrow zone that stretches in the direction of NW-SE sediments of Upper Devonian and Lower Carboniferous appear. Variscan structural stages are represented here with distal flysch facies: argilic and argilophyllite politic rocks periodically interfinering with thin layers of fine-grained, seldom with coarse-grained sandstone, turbiditesand and occasionally debris flow deposits. There are also sericitic and chlorite schist and very rarely black lidytes and lenses and clasts of limestone. East of Niška Banja, along river Nišava, Permian deposits are developed. They are represented with red beds of typical "new red sandstone" facies formation.

Upper Jurassic (Tithonian) in a more than a few places comes up to the surface in Miocene sediments. It is presented with sub reef and reef limestones or shallow water algal-foraminoferal limestones and dolomites.

Lower Cretaceous (Aptian) in the direct vicinity of Niška Banja is represented with sandstone, marl and limestones and further to the south prevail conglomerate, sandstone, aleurolite and limestones of Urgonian facies. This unit is of considerable importance considering the fact that on its contact with Quaternary spring sediments, thermal springs of Niška Banja occur.

Miocene (Middle and Upper) stretch in the east part of Niš basin. They overlapping lower Miocene coal series or the older rocks. Miocene sediments include clastic sediments represented with clays with coal seams, sands and gravel.

Quaternary sediments in the direct vicinity of Niška Banja considered as alluvium and spring sediments. Alluvium sediments are represented with gravel, sand and clays of small thickness. Spring sediments of Niška Banja contain carbonate travertine.

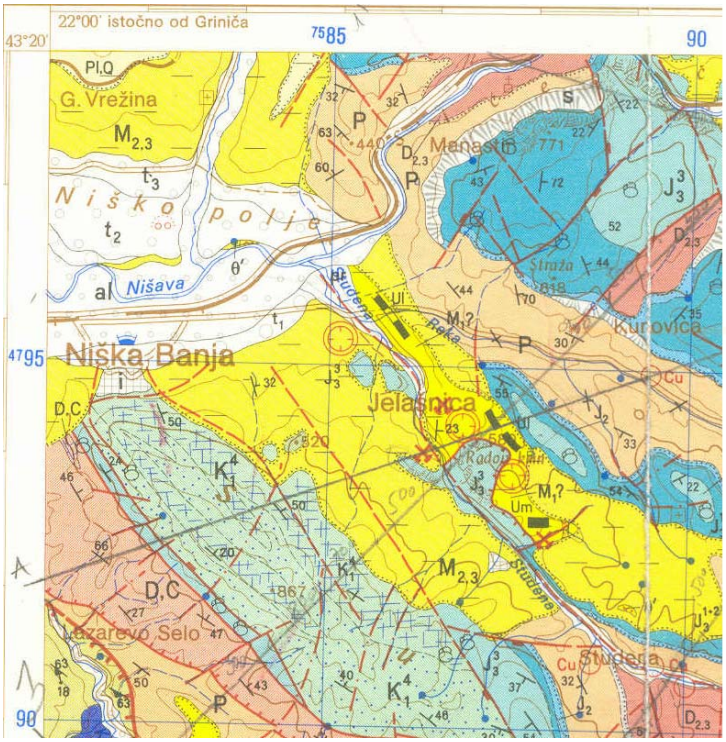

Figure 3.1 *Section of geological map showing area around Niška Banja*

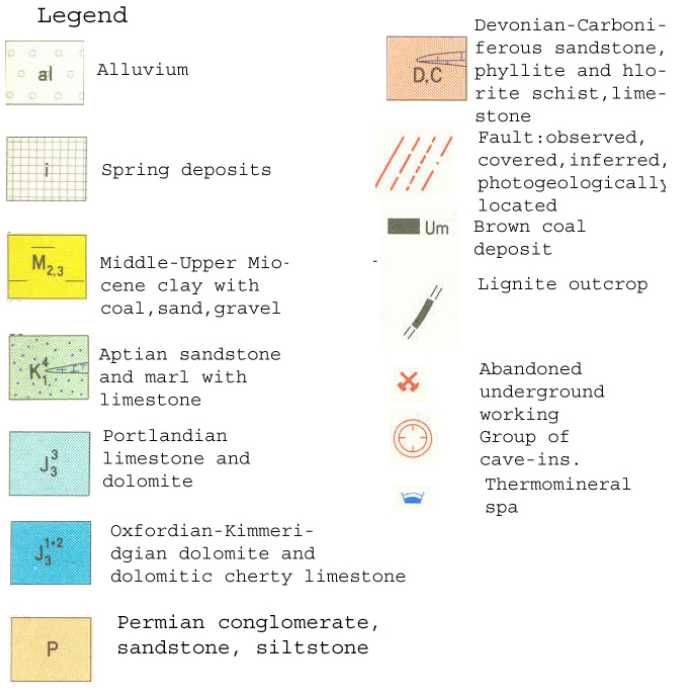

Figure 3.2 *Legend for geological map*

A particularly interesting formation is the quaternary travertine, spring sediment formed as a result of carbonate precipitation from spring waters of Niška Banja. Generally the travertin is formed when water saturated with carbonate runs out from the spring, pressure and temperature drop down, and the solubility of carbonate decreases. In this moment carbonate precipitate and travertine is formed. A travertine plateau occupies central parts of Niška Banja.

During works a long time ago, travertine was cut deeply so it is quite easy to see that travertine is not uniform in terms of porosity. In some places we have parties of almost compact, lithified travertine. However, little further away, there are parts of strongly porous travertine of barrier/waterfall faces, with hollows, frequently molds after decayed wood logs and branches, channels and caverns. Interbeds, lenses and pockets in travertine filled with clay, detrital travertine gravels and sands with terrestrial gastropods, marl and clay with marl, generally as sediments of "pools" are seldom. Until now it is known that travertine in Niška Banja is precipitated from the waters of Glavno Vrelo spring. It is estimated that the speed of travertine sedimentation is 0.8 mm per year, and the age of basal, oldest travertine bed is 27 000 years.

Geology of Niška Banja basement can be best viewed through a drill hole, made during period 1986 to 1987. Drill hole is located in the southern part of Niška Banja town. In this way we can have a good look into the geological structure of Niška Banja. Starting from surface and going downward, composite summary geological profile under the town is such as follows:

0-135 m, clay and marl sediments – Neocene
135-220 m, flysch sediments – Lower Cretaceous
220-350 m, Lower cretaceous limestones with abundance of clay
350-470 m, compact lower Cretaceous limestones
470-510 m, limestones with mineral water

The central parts of the Niška Banja town are covered with the formation of travertine. Travertine shows a high level of natural radioactivity.

4 Description of measurement methods and techniques

A good quality of measurements implies calibration of all used detectors and instruments. In other words, this means that the sensitivity of the detectors has been checked, compared, and verified. It is also necessary to repeat periodically calibration of the detectors and participate in inter-comparison measurements.

In order to check the sensitivity of detectors, i.e. verify the accuracy of measurements, inter-calibrations of detectors are performed. This means that the detectors are exposed to an "unknown" but identical radon concentration in a series of exposures. In this way the calibration coefficient is determined for each detector for the given radon concentration [Goldin, 1984]. These inter-calibration series are performed periodically. Inter-calibration is usually carried out by using a "radon chamber" where the parameters are controlled, i.e. the amount of radon and temperature, humidity, speed of air flow, size of aerosol particles, etc. In general, it can be concluded that the calibration of detectors for radon progenies is more difficult to perform compared to calibration of radon detectors, one of the many reasons being that there is no standard for radon progenies [NCRP, 1988; Nazaroff, 1988]

4.1 Contemporary radon and gamma measurements in dwellings

The common practice in residential studies has been to measure contemporary radon concentrations, asthez are at the time of measurment. Such "actual" measurements often consist of the averages of short-term radon measurements. These are often extrapolated to longer time periods. This can lead to very large errors in the estimation of the radon exposure which can introduce corresponding errors in risk factors derived in epidemiological studies. The use of a 12-month radon-monitoring period reduces errors associated with the application of average seasonal adjustment factors applied to short-term radon measurements in individual residences.

Lagarde [Lagarde, 1997] stated that errors in the exposure assessment should be taken into account in risk estimates based on residential studies, and suggested that the scope of errors in risk estimation be broadened. In addition, the influence of thoron interference in radon measurements and hence risk estimation requires further evaluation [McLaughlin and Žunic, 2005; Lagarde, 2001].

Based on the duration of sampling there are several different methods of measuring radon and radon progeny concentrations so that Contemporary Radon Gas Measurement Techniques can be divided into three categories and these can be classified as (1) instantaneous "Grab Sampling", (2) integrating "Time Integrating Sampling (3) continuous recording "Continuous sampling".

Integrating measurement -"Time Integrating sampling" provide a mean value of the radon content or radon progeny content over a defined measurement period, which may last from a few days up to several months or a year (for example, by charcoal, track etch film or electret-based material, the latter is a charged material that will be gradually discharge by the alpha particles from radon and progeny and Thermo-luminescence dosimeter (TLD). Integrating detectors provide in fact a single long-term radon gas concentration measurement which makes them ideal for survey work or for use to determine the annual average concentrations. Integrated radon gas measurements for this work were made using a passive diffusion chamber, initially designed by the National Radiological Protection Board (NRPB) of the UK

and modified by SSI (Swedish Radiological Protection Institute) and later modified by the National Institute for Radiological Sciences, (NIRS), Chiba, Japan [Doi et al., 1992].

4.1.1 CR-39 solid state nuclear track detectors (SSNTD)

The most commonly used detectors for the passive detection of radon are known as Solid State Nuclear Track Detectors (SSNTD) or as track-etch detectors. The most suitable and most common for indoor measurements is CR-39, a polymer derived from the monomer oxydi-2, 1-ethanediyl di-2-propenyl diester of carbonic acid [Cartwright et al., 1978; Cassou and Benton, 1978].

CR stands for "Colombia Resin" and its physical appearance is similar to Perspex. It was shown that it was a particularly sensitive etched track detector [Cartwright et al., 1978] The optical quality and the response to incident radiation of the CR-39 depends on the initiator and the additives during the manufacturing process. A small quantity of a plasticizer D.O.P (di-iso-octyl-pthalte) or D.N.P. (di–nonyl-phthalte) is added during manufacture and this improves the quality of the post-etch surface and, more importantly, alters the response of the bulk plastic material to alpha particles. Addition of D.O.P improves the optical clarity and reduces the number of surface defects, while D.N.P. improves the homogenity of the CR-39 but decreases the etch rate. CR-39 is sensitive to alpha particles from energies of 0.1 MeV up to 60 MeV [Cartwright et al., 1978; Fleischer et al., 1978] proposed that track formation is dependent on the total energy deposited per unit path length by the incident ion. When an alpha particle strikes and penetrates CR -39, it slows down to rest by depositing its energy in a series of interactions with the surrounding material. The energy loss per unit distance travelled (dE/dx) is referred to as the stopping power. Tracks are formed when the Linear Energy Transfer (LET) expressed as dE/dx, the energy loss per unit distance travelled, exceeds some critical value. This can be calculated using a form of the Bethe-Bloch equation [ICRU, 1993]. Alpha particles penetrating the material cause damage to the chemical bonds. After chemical etching, the damaged regions become visible when magnified and are seen as conical tracks or holes in the plastic.

It consists of a two part polypropylene conducting holder and a CR-39 detecting element (13 mm x 37 mm). The detector shell is impregnated with carbon fiber and therefore is conducting to avoid any electrostatic effects that might affect the behavior of the charged radon progeny inside the chamber. The upper part of the detector is hemi-spherical and has a circular retaining strut that holds the CR-39 element in place in a recess in the base. Radon gas enters the detector chamber by diffusion with a half-time of about 25 minutes [Gilvin and Bartlett, 1988] which is short in comparison to the half-life of Rn-222 of 3.8232 days (Walen and Bastin 1959). It has been shown that the long term average radon concentrations inside the holders is essentially the same as the long term average concentration outside, though there may be short-term variations in the outside concentrations [Wilkenson and Saunders, 1985]

A protocol for the preparation of CR-39 detectors was developed, which included the anti-static precaution of dipping the plastic in a 1:500 solution of detergent and water which has been shown to reduce the static charge on the plastic [Fitzgerald, 1994]. The holder acts as a simple radon diffusion chamber, excluding radon decay products and dust, limiting access of moisture but allowing the entry of radon gas [Bartlett and Bird, 1987].

4.1.2 LR 115 solid state nuclear track detector (SSNTDs)

Another SSNTD used frequently in this work is LR 115, a cellulose –nitrate film produced by Kodak Pathe. This film consists of a thin (13 micrometers), red alpha particle sensitive layer and a supporting transparent film. LR 115 has a much narrower energy "window" than CR-39 (a property that made it very useful for certain aspects of the work described here). This energy window is highly sensitive to etching

conditions, and for etching conditions of a 70 minutes etching in 2.25 N NaOH at 60 ^{0}C, the energy window is approximately 1.2-2.4 MeV. This detector is easily charged which may disturb the radon progeny behavior near the detector. Thus great care must be taken to reduce or eliminate the static charge, by dipping the LR 115 in a dilute (1:500) solution of detergent and then by allowing it to dry before use. The LR 115 film is also very susceptible to damage while handling, as the sensitive layer is so thin (13 micrometers) it is easily removed. Gloves were worn at all times when handling the film as even grease from handling can affect the etching. LR 115 is also sensitive to damage by ultra-violet radiation, such as sunlight. After exposure to UV light the sensitive layer disintegrates during etching and accurate counting is impossible. When counting the tracks in LR 115 using a microscope a light filter is used which creates a quasi – monochromatic light source at a wavelength of 546 nm, since both optical lenses and the human eye respond best to green light.

4.1.3 Thermoluminiscent detector – LiF: Mg, Cu, P (MCP-N detectors)

The environmental dosimeter consists of a KD-85 dosimetry card containing three highly sensitive MCP-N (LiF: Mg, Cu, P) detectors closed in a 2 mm thick (PVC) container, packed in a light-tight aluminum foil and a water - proof polyethylene bag. The package is mounted on a steel rod at the height of 1 m above ground level. For read out, the ACARD97 automatic TL reader (Microlab, Poland) is used. Detectors are annealed in the PTW-TLDO programma ble furnace. The environmental dosimeters are calibrated free in air, in terms of kerma in air, using the Cs-137 beam at the Laboratory for Calibration of Radiation Protection Instruments at the Institute for Nuclear Physics, Krakow, Poland.

The main parameters of MCP-N such as fading, light sensitivity, Lowest detectable Dose, LD, self-dose, zero-dose, energy response from low-energy X-rays up to 6-7 MeV, influence of annealing and read - out conditions on detector stability, were investigated and optimized [Ross Hirning, 1992; Saez-Vergara et al., 1996; Budzanowski et al., 1996; Budzanowski et al., 1997; Budzanowski et al., 1998; Budzanowski et al., 1999; Budzanowski et al., 2000; Dugganet al., 2000]. The values of these parameters, against the ones that refer to standard LiF: Mg, Ti (MTS-N, TLD-100 equivalent) detectors, are listed in Table 4.1 (Olko et al., 2004). The golden rule for handling LiF : Mg, Cu, P detectors is never to heat them above 240 °C, as this can lead to a decrease of detector sensitivity. Therefore, during read out it is recommended either to apply step heating up to 240 °C or linear heating up to about 260 -270 °C, but for a limited time of a few seconds only. The recommended annealing procedure consists of heating for 10 minutes at 240 °C, followed by rapid cooling on a thick aluminum block, at room temperature. TLD reader annealing at 240 °C is also possible. After their irradiation detectors are additionally annealed for 10 minutes at 100 ^{0}C to remove the low-temperature peaks.

***Table 4.1**. Main parameteres of MCP-N detectors (Olko et al., 2004)*

Parameter	Reference	Value
Sensitivity (as compared to TLD-100)		25 times higher
Lowest dwtectable dose, L_D	Budzanowski, M., 2000. Ross Hirning, C., 1992.	60 nGy
Linearity range	Budzanowski at al, 1997.	from 200 nGy to 5 Gy
Self dose	Budzanowski at al, 1996.	less than 1 $nGyh^{-1}$
Fading	Budzanowski at al, 1998.	less than 5% per year
Sensitivity to light	Duggan at al, 2000.	negligible
Energy response (60 keV to 6-7 MeV)	Seaz-Vergara, 1996.	< 20%
Zero-dose	Budzanowski, M., 2000.	150 nGy
Time estimation using peak ratios	Budzanowski at al, 1999.	up to 3 months

4.2 Retrospective measurement of radon exposure

Retrospective assessment of radon exposure is a technique which was developed in the past twenty years [Oberstedt and Vanmarcke, 1996; Samuelsson, 1988].

Retrospective methods are based on measuring ^{210}Pb or ^{210}Po due to radon which has accumulated in solid or porous media over many years and which is therefore in principle proportional to the cumulative radon exposure.

Basically two methods of retrospective radon assessment exist. These methods are based on the buildup of low levels of ^{210}Pb and consequently ^{210}Po activity that can be related to the long term average radon concentration.

In the surface trap technique, the airborne radon decay products deposit on smooth glass surfaces usually those found in photograph and picture frames, where subsequently , due to the decay of these deposited progeny, their decay products can be implanted through alpha particle recoil, and remain fixed in the near surface layer of the glass, where they can be detected through alpha decay by means of track etch detectors or pulse ionization chambers [Falk et al., 1996; Lively and Steck 1993; Samuelsson et al., 1992; Samuelsson, 1988]. These are often easy to date simply by asking the owner about the age of the glass. Glass surfaces of other objects, such as mirrors, glass doors between rooms and clocks, have also been used. Windowpanes are usually avoided as the airflow pattern and therefore the deposition of progenies near windows is not typical, and UV-radiation influences response of the detector materials.

The use of glass in epidemiological studies is now becoming more common. Studies involving measurements on glass have been performed beside Serbia in USA, Norway, Germany, Ireland and Sweden

[Zunic et al., 2007; Paridaens et al., 2001; Falk et al., 2001; Zunic et al., 2001; Birovljev, 2001; Cauwels and Poffijn, 2000; Hamel, 1999; Walsh, 1999; Mahaffey et al., 1999; Alavanja, 1999; Field et al., 1999; Palfalvi et al., 1995; Mahaffey et al., 1993] .

The second retrospective technique used in this work is a volume trap technique [Oberstedt and Vanmarcke, 1996]. In this case, it is the radon gas itself which diffuses freely into bulky, porous materials, depositing there its decay products. Thus, the short lived decay products are trapped in the volume trap, and give rise to a build-up of ^{210}Pb and ^{210}Po. After about 18 months, equilibrium is reached between ^{210}Pb and ^{210}Po which is an alpha emitter (5.3 MeV) with a half life of 138,3763 days (Hayward R.W., et al., 1955). It is this isotope that has to be separated for analysis by radiochemical means. The volume of the sample has to be determined and it can be done by weighing if the density is known, or less precisely by cutting sample in a shape that fits into a receptacle l of known volume. It is important that the sample should not have been contaminated with airborne decay products during its lifetime, hence, the need to first remove the surface of the volume trap.

Volume traps are a conceptually similar technique to surface traps, i.e. where the ingression of ^{210}Po from ^{222}Rn which has diffused into the interstitial surfaces of porous household materials has taken place. The main advantage of volume traps such as mattresses, cushions and spongy household materials is that they monitor the radon concentration directly, and are thus unlike the surface traps independent of aerosol conditions in the surrounding air , which are influenced by complex processes such as turbulence or air movement. As the analyses, however requires radiochemical separation the sample will thus be destroyed [Oberstedt and Vanmarck, 1996]. In practice, a small sample of the material is taken from the center of the volume trap. For polyether volume traps, samples with a weight between 0.5 g and 3 g are usually taken. Any bulky, sufficiently porous material, with known age and history, can serve as a volume trap. The material should, however, be such that radon decay products cannot migrate in to or out of the volume trap. Moreover, the volume trap material should not contain a significant natural ^{210}Po background, so as to disturb the measurement results.

4.2.1 Surface trap “CR-LR difference technique”

A technique using CR-39 (polyallydiglycol carbonate) and LR 115 detectors (cellulose nitrate) “CR-LR difference technique” aiming at large-scale fieldwork was used in the work described here to measure ^{210}Po [Falk et al., 1996]. The alpha sensitivity of CR-39 (plastic material), which is the same type of material typically used in ambient radon detectors enables the activity measurements on glass using an alpha track-etch detector and also on the other hand to fulfill the need for a practical and economical way of estimating long term radon concentration. This method allows the measurement to be made without removing the glass from the home. Another advantage is that such detectors are passive and require no power supply. In this technique a few square centimeters of each of the alpha track detectors are mounted side by side on a chosen flat glass surface. The glass is cleaned, and a piece of CR-39 plastic is fixed to its surface and left in place for a few weeks. For standard etching conditions the LR 115 is sensitive to alpha particles in an energy window in the approximate range 1.2 – 4.8 MeV, while the CR-39 is sensitive to alpha particles with energies from less than 1 MeV to much greater than the maximum energy (7.68 MeV) emitted by radon progeny. As the ^{210}Po alpha energy is 5.3 MeV the LR 115 will not record tracks from the surface trapped ^{210}Po, however will produce tracks proportional to the intrinsic alpha activity naturally present in the glass. The CR-39 track density will thus be a mixture of tracks due to the implanted ^{210} Po, alpha emissions and those from the intrinsic alpha activity of the glass. After being deployed for T hours the following expression gives the surface ^{210}Po activity for a glass surface onto which side by side CR-39 and LR 115 detectors were mounted:

$P = (CR - B \times LR) / (T \times K)$ (1),where P = ^{210} Po activity in $Bq \cdot m^{-2}$, CR =CR-39 net tracks per cm^2, LR =LR 115 net tracks per cm^2, B=CR-39:LR 115 intrinsic alpha activity track density ratio for unexposed glass and K= PIC (Pulse Ionization Chamber) determined CR-39 sensitivity factor to ^{210} Po. B and K values depend on the actual etching regimes and track acceptance criteria used. For standard procedures

values of B=1.97 and K=0.081 tracks cm^{-2} / $Bq \cdot m^{-2}$ hr were obtained [Falk et al., 1996].

The typical exposure period used in this work for the alpha track detectors mounted on glass was about three to four months. The plastic detectors are then removed and the alpha tracks resulting from implanted ^{210}Po into its surface , that have been etched, are counted. The track generation rate is then a measure of the alpha-activity of the glass surface. Using the measured ^{210}Po value together with a modified Jacobi radon progeny room model the mean radon concentration in a room over the many years the glass was exposed can be calculated. The radon concentrations obtained in this way, correlated to the radon decay products, can be used to create radon exposure histories [Falk 2001, McLaughlin 1998, Falk 1996]. The CR-39 alpha track densities recorded for these exposures were mainly within the range 5 to 25 tracks mm^{-2}. Measured ^{210}Po values ranged from about 3 to 30 $Bq \cdot m^{-2}$ with individual values as high as 50 $Bq \cdot m^{-2}$. Contemporary air indoor radon gas concentration were measured in the chosen dwellings by means of standard passive diffusion type CR-39 radon detectors with a calibrated radon sensitivity of 2.6 alpha tracks cm^{-2} / $kBq \cdot m^{-3}$ h.

4.2.2 The volume trap technique

The volume trap measurement technique as being mentioned above is an alternative retrospective radon measurement technique to the surface trap measurement technique described in this work but it also utilises the measurement of ^{210}Po surface activity arising from the decay of the long-lived radon progeny ^{210}Pb. However, in this case the ^{210}Po activity is not measured on a glass surface or mirror. Instead, the measurement is carried out to determine the ^{210}Po activity that has built up or ingrown in spongy materials such as mattresses or cushions. This idea was first proposed by Samuelsson and Johansson, 1994.
These spongy materials allow the diffusion of radon gas through its pores. In normal circumstances, radon progeny cannot diffuse easily into the material and consequently the progeny deposited at depth in the material is attributed entirely to the decay of diffused radon gas itself. Thus, the ^{210}Po activity in the centre of the material correlates well with the radon gas concentration in the room air where it has been exposed to over its lifetime allowing a retrospective radon gas measurement.

The measurement material for the volume trap technique is typically a 100 cm^3 polyester foam sample taken from mattresses or cushions used in a dwelling. In order to exclude any surface effects, the sample is normally dissected from the inside of the exposed material. Once the sample has been removed from the material it is kept in a radon free environment for approximately 138 days in order to reach radioactive equilibrium. The next stage of the process is to chemically separate the ^{210}Po from the material. This is done via several steps [Narita et al., 1989, Benoit and Hemond, 1988].

Firstly, the samples are dissolved in NaOH (14M) and hydrolyzed for 5 to 10 hours. After cooling, HCl (8M) is added and the solution is dried under infra-red lights. This residue is then dissolved in HCl (12M) in order to eliminate all NaOH. The final residue is then dissolved in 4.2 ml of HCl (12M) and the sample volume is adjusted to be 100 ml using water. This solution is mixed with 100 mg of ascorbine acid and is covered with the silver plate. And the apparatus is turned around to place the silver plate at the bottom to let the polonium auto deposit onto the plate for 48 hours. The activity of ^{210}Po is then determined via alpha-partricle spectrometry using a PIPS detector. The amount of ^{210}Po lost during this procedure is determined by using ^{208}Po as a tracer material.

The retrospective radon gas concentration is estimated from this ^{210}Po activity using a conversion factor that takes into account the porosity and the age of the sample material. These detectors have been used in surveys in the past [Birovljev et al., 2001, Paridaens et al., 2001; Paridaens et al., 2000] found the measurement technique to be very reliable.

However, like all radon measurement techniques it has some disadvantages. One of the major disadvantages is that the technique is destructive (i.e. the material is removed from the dwelling and

destroyed during analysis unlike the surface trap technique) and so occupants are sometimes reluctant to supply samples for the measurement. In addition, it has been found that in very dusty circumstances direct penetration of radon progeny from the outside of a sample to the centre can be possible and so extra caution is needed in these circumstances.

5 Experimental investigation of radon

5.1 Inter-comparison of radon detector sensitivities (calibration coefficients)

The calibration coefficient of a track-etched detector is function of the procedure of etching and counting traces. Calibration coefficients are determined through participation of the corresponding laboratories in international inter-comparisons of detector sensitivities, held usually at the National Radiological Protection Board (NRPB, UK) in Oxford. In this work inter-calibration is carried out before each field work so that it could be performed using standardized detectors of known sensitivities. In the course of 1997, 1998, and 1999 three inter-calibrations of sensitivities of detectors used in this experimental work have been done through participation in international inter-calibrations at the National Radiological Protection Board (UK) [Naismith et al., 1998; Howart and Miles, 2000]. The calibration factor was 2-3 traces $cm^{-2}/kBq \cdot m^{-}3 \cdot h$. Table 5.1 shows individual values of the calibration coefficient obtained at the three inter-comparisons of detector sensitivities.

Table 5.1 *International inter-calibration of sensitivities of solid state detectors of charged particles*

No	*Place*	*Number of participating laboratories*	*Year*	*Number of sets of detectors*	*Number of detectors per lab.*	*Type of detectors*	*Calibration coefficient*
1	*NRPB*	*62*	*1997*	*87*	*40*	*CR-39 (PADC)*	$2.6tr \cdot cm^{-2}/kBq \cdot m^{-3}h$
2	*NRPB*	*54*	*1998*	*95*	*40*	*CR-39 (PADC)*	$2.7tr \cdot cm^{-2}/kBq \cdot m^{-3}h$
3	*NRPB*	*54*	*1999*	*193*	*40*	*CR-39 (PADC)*	$2.7tr \cdot cm^{-2}/kBq \cdot m^{-3}h$

tr - trace

5.2 Organization of field work

5.2.1 Selection of locations

On the basis of data on uranium prospecting in the area of Kalna (former uranium mine open 1948, closed 1966), and Gornja Stubla [Jakupi et al., 1997], geo-chemical approach has been adopted for selecting these two field locations. In the case of Niška Banja, being the spa, Vinča team was invited for supervision of the indoor radon measurements done 2000 [Manić at al., 2006]. After this activity the field work was continued in Niška Banja which was chosen as the third field location on the base on its water radium high content.

5.2.2 Questionnaires and work lists for field work

5.2.2.1 Questionnaire for measurement of radon concentration

This questionnaire is made according to the questionnaires used for performing national programs for radon in Ireland and Germany[1] and adjusted to the conditions of living in Serbia (former Serbia and Montenegro). Data contained by this questionnaire concern: code number of detector, starting and ending times of exposure of detectors placed in closed places to measure radon concentration.

Particular attention has been paid to data concerning the characteristics of closed spaces: type of foundation on which the house is erected, time of building, building material, type of floor, type of water supply, floor of the closed space, existence of hydro-isolation, type of heating, type of ventilation, number and age of inhabitants.

5.2.2.2 Questionnaire for measurement of thoron concentration

The questionnaire is made on the basis of the questionnaire[2] used for performing national program for radon in Japan and contains information significant for measurement of thoron (and radon). The most important entries concern the age of house, building structure, closed space where detector is installed (height and distance of the detector from wall), floor of the closed space, type and size of the closed space and surface material of the walls.

5.2.2.3 Protocol regarding variations and seasonal exposure

The list is made for the purpose of performing field work. It contains location of indoor spaces (house) for the map, name of the owner of the house, the time periods of the detector exchanges (exposure time in days) with additional two columns, one for code of detector and the another one for measured concentration.
On the basis of this list the concept for making maps for detector locations is made and it enabled data processing concerning not only of the measured average annual concentrations but they contained data used for application of the model of seasonal variations of radon.

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[1] **The questionnaires from Ireland and Germany were obtained by courtesy of Prof. James McLaughlin, University College, Dublin, Ireland.**

[2] **The questionnaire from Japan was obtained by courtesy of Dr Kenzo Fujimoto, Chiba, Japan.**

5.2.2.4 Protocol list for measurement of indoor and outdoor gamma dose rate

The work list contains, as the previous one, name of the owner of the closed space, type of the closed space, and entry for recording the number of measured pulses (environmental meter), and period of exposure. Regarding TLD dosimeters it was written each card number, date of start end end of exposure and results of gamma dose rate in nGy per hour.

5.2.2.5 Questionnaire for retrospective measurements

The questionnaires used in the selected rural communities have been obtained from Ireland and Sweden. In addition to Ireland (University College Dublin - UCD), identical questionnaires for their detectors have been used by investigators from Norway (National Radiation Protection Association - NRPA, Oslo), Italy (Centro Radiazione Recerca - CRR, Verona) and Sweden (Swedish Stocholm Institute – SSI). Detectors have been sent together with the questionnaires in order to carry out inter-comparison SINI (Sweden, Ireland, Norway, Italy) at the zones of Kalna and Gornja Stubla. The problem here was measurement of activity of the long lived radon progeny ^{210}Po whose activity was monitored by using glass ("surface traps" method) for the purpose of determining the stability of these zones and the exposure of the population involved. The important data contained by the questionnaire are: type of the closed space, type of glass involved, date of inserting detector, and code number of detector. For the other retrospective method ("volume traps") the questionnaires for radon have been used because it was necessary to determine only the age of the sample, marked as an additional date to the radon questionnaire.

5.2.3 Health questionnaire

For Gornja Stubla field location the health questionnaire was made as an original initiative aimed at obtaining the preliminary data, the so called anamnesis concerning the length of stay of inhabitants in the examined homes, indirectly migrations of the population, type of malign diseases or other diseases characteristic for the examined locations in particular tuberculosis and bronchial asthma, and data on professions and length of service of the individuals involved.

5.3 Field work in the rural communities

For the purpose of obtaining realistic values of average annual radon concentration in the selected houses, series of detectors were deployed continually in one or more of rooms (bedrooms and living rooms) where the people live or sleep during a period of 12 months. Each detector was exposed in the chosen room for about 3 months and then replaced by a new detector. In this way the total of four such detectors per each selected place were used contributing in this way to obtaining seasonal and average annual value of radon concentration. In a small number of cases, detector was not replaced after the exposure period, mainly because of the absence of the owner, and the detector was left for the next period of exposure or until the detector was finally removed.

5.3.1 Rural community of Kalna

In the zone of Kalna, having 500 of inhabitants, during 1997/1998, in four periods of exposure (Table 5.2), a total of 406 detectors of Swedish design (SII/NRPB), of type CR-39, were used in the selected rooms at eleven locations: Inovo (K1), Kalna (K2), Stara Kalna (K3), Belevica (K4), Mezdreja (K5), Ravno Bučje (K6), Janja (K7), Gabrovnica (K8), Donje Polje (K9), Balta Berilovac (K10), Vrtovac (K11). In an area 40km x 40km, in 75 houses there were 103 measuring sites, 74 in living rooms and 29 in bedrooms. A total of 98.7% of detectors have been changed and developed. Seasonal exposure of solid state alpha track detectors (CR-39) included four seasons (Table 5.2) and 11 locations.

Table 5.2 *Exposure and changes of CR-39 detectors in closed spaces in Kalna*

Change of detector	*Period of exposure*	*Number of days*
I	*08.03.1997 - 06.07.1997*	*120*
II	*06.07.1997 - 23.10.1997*	*109*
III	*23.10.1997 - 13.02.1998*	*72*
IV	*13.02.1998 - 05.06.1998*	*112*

5.3.2 Rural community of Gornja Stubla

In Gornja Stubla, 3000 inhabitants, a total of 709 detectors in 65 houses situated in an area of 3km x 2km were exposed during period 21.04.1997 - 20.02.1999.
A total of 89% of detectors has been replaced, whereas 12 % is lost exposed owing to the war situation.

5.3.3 Rural community of Niška Banja

Radon survey has been carred out around the town of Niška Banja in region partly located over travertin formation, showing an enhanced level of natural radioactivity. Indoor radon concentration were measured using CR-39 diffusion type radon detectors in 102 living rooms and 102 bedrooms of 65 family houses (contemporary radon as well as both retrospective methods: surface trap and volume trap). Outdoor radon measurements were performed at 56 points evenly distributed over the teritory of Niška Banja (in the

gardens, 1 m abowe soil, 2-7 m away from the house wall). All selected houses are single-family, one-or two- flore buildings. The outdoor and indoor measuremenst were conducted over the period March 2003 to January 2006 during different seasons in order to account for seasonal variation of radon concentration. Additionaly outdoor and indoor gamma dose rate were measuremed in terms of kerma in air, with high-sensitive LiF:Mg, Cu, P thermoluminescence detectors. TL dosimeters were exposed in 55 outdoor and 55 indoor location during four mounths over the period November 2003 to April 2005.

6 Results

A. Identification

As decribed in the 11 published scientific papers, over the period 1999 – 2009, which are the main body of the thesis, a series of field investigations of the population radon exposures into predominantly rural communities were carried out. Of particular interest was the identification and assessment of the exposure of the general population to high levels of indoor radon at locations in Serbia, i.e., Kalna (Southeast Serbia), Gornja Stubla and Niška Banja (Southern Serbia).

In addition to making

- *contemporary radon measurements using nuclear track detectors [Paper III, Paper VI, Paper VII] for the first time in this region*
- *retrospective radon assessment methods and techniques were used to determine radon levels in past decades [Papers I – Surface Trap(ST), Paper II – Volume Traps (VT) , Paper III- ST and VT , Paper IV, Paper V –ST and VT, Paper VIII – ST and VT].*

Furthermore, also for the first time in this region,

- *thoron measurements [Paper III, Paper XI], were made in a number of field locations using Japanese passive discriminative polycarbonate [Paper III] as well as Japanese CR 39 passive discriminative nuclear track detectors [Paper XI]. Thus, this was an innovation in this region. Indoor radon levels in excess of 6 kBq·m^{-3} were found to be present in two, out of three, investigated areas namely: Niška Banja and Gornja Stubla, where, particularly, in addition indoor thoron levels was found to be present more than 1.5 kBq·m^{-3}.*
- *The geological/geochemical origin [Paper IX, Paper XI] of these high radon levels was identified in this work showing that the entry of soil gas into the dwellings is the main contributor to indoor radon levels.*
- *The radon /thoron soil gases examinations was advanced and thus more detailed reserach [Paper IX, Paper XI) and were also provided at some field locations (Niška Banja) for the first time as well the*
- *comparison of their distributions at low and high background area (Niška Banja and Thermopower plant surroundings TENT B, Obrenovac near Beograd) [Paper XI].*

Quite challenging were

- *examination of the thermomineral water samples on radium content [Paper X].*

For the most of the field sites dealing with indoor radon investigations.

- *Outdoor radon and gamma activities (Paper III, Paper VII, Paper XI) were obtained..*

All results gave the opportunity for

- *the comparison of the methods and techniques (Paper XI) as well as the field areas which were thoroughly presented*
- *at two international workshops, i.e., the first one:*

  (ECE I): "Promotion of New Electrochemical Etching Facility (ECE) and Its Application to Natural Radiation Studies in Western Balkan Countries", Belgrade, June 30 to July 3, 2003, Hotel Trim-Kosutnjak and Vinca Institute, ISBN 86-7306-060-5 Eds Z.S.Zunic, K.Fujimoto and the

second one:

(ECE II): "The second Vinca ECE Lab advanced research international workshop – the new perspectives for thoron survey and dosimetry", Niška Banja, June 6 -10, 2005, Serbia and Montenegro, ISBN 86-7306-069-9, eds Z.S.Zunic, K.Fujimoto.

The all findings have formed the basis for currently establishment of radon risk map over Serbia being the part of potential European radon atlas.

6.1 Paper resumes

PAPER I - Resume

The use of SSNTDS in the retrospective assessment of radon exposure in high radon rural communities in Yugoslavia

Radiation Measurements, Volume 31, Issues 1-6, June 1999, 343-346

Z. S. Zunic, J. P. Mc Laughlin, C. Walsh and R. Benderac

In most radon epidemiological studies contemporary radon measurements made in the present dwellings of the case or control subjects have been used as surrogates for concentrations in past decades even though changes in radon levels and residence may have occurred.

Better knowledge of the radon levels in the past if available, would improve the estimation of long term radon exposures of the subjects and hence a better estimation of radon risk would be obtained. Thus, radon induced lung cancer risk estimates should ideally be based on estimated long term cumulative radon exposure rather than on the less reliable contemporary radon concentration. Taking into account the long latency period for lung cancer induction, it is clear that the contemporary or present level of radon a person is exposed to is not of great relevance to their cumulative risk to date. In recent years retrospective assessment of radon exposures has become possible by the use of techniques which measure the build-up of the long-lived radon progeny ^{210} Po "trapped" in the surface layer of glass objects that have been in a house for a known number of years. This is known as the "surface trap" retrospective technique to distinguish it from the "volume trap" technique (see following paper). Using a modified Jacobi room model, referred to usually as the "room model", which takes into account radon progeny behavior in enclosed spaces like rooms, the mean radon level in the past can be estimated.

In this paper a description is given of field work in which retrospective and contemporary radon measurements were carried out in two small rural communities in Yugoslavia known from previous field investigations and geological characteristics to be areas of elevated natural radiation. These are Kalna in southeast Serbia and Gornja Stubla in Kosovo. In both these areas, elevated concentrations of uranium and thorium are present in rocks and soil.

Large-scale fieldwork, using the "CR-LR difference technique", developed by the Swedish Stockholm Institute (SSI) in Stockholm and University College Dublin in Dublin, using alpha track detectors was used in the work described here to measure ^{210} Po in glass surfaces (Falk et al.,1996). In this technique, a few square centimeters of each of the alpha track detectors CR-39 (polyallydiglycol carbonate) and LR 115 (cellulose nitrate) are mounted side by side on a chosen flat glass surface. For standard etching conditions the LR 115 is sensitive to alpha particles in an energy window in the approximate range 1.2 - 4.8 MeV while the CR-39 is sensitive to alpha-particles with energies from less than 1 MeV to much greater than the maximum energy (7.68 MeV) emitted by radon progeny. As the ^{210}Po alpha energy is 5.3 MeV the LR 115 will not record tracks from the surface trapped ^{210} Po but will produce tracks proportional to the intrinsic alpha activity naturally present in the glass. Defining the quantity R as being the ratio between the contemporary and the room model estimated retrospective radon concentrations it was found that the majority of R values for the dwellings investigated lay within a factor of three of the R=1 value. This is taken as a good indication that there has been no significant change in mean radon levels in these two communities over recent past decades and these areas of Yugoslavia may therefore be considered as suitable potential radon epidemiological study locations.

In this paper an account is given of the principles and some field applications of the surface trap retrospective technique. The alpha track detectors CR-39 and LR 115, are used to measure alpha recoil implanted ^{210} Po in glass surfaces (surface traps) by means of which the cumulative radon exposure in a dwelling in past decades may be estimated using a modified Jacobi room model. Thus, radon induced lung cancer risk estimates may be based on estimated long term cumulative radon exposure rather than on the less reliable contemporary radon concentration. Taking into account the long latency period for lung cancer induction, it is clear that the contemporary or present level of radon a person is exposed to is almost irrelevant to their cumulative risk to date. It should be noted that this was the first time that retrospective radon assessment techniques were used in any part of the Balkans.

PAPER II - Resume

Retrospective Radon assessment by means of ^{210}Po activity measurements

Applied Radiation and Isotopes, Volume 53, Iss 1-2 (2000) 361-364

J. Paridaens, H. Vanmarcke, K. Jacobs and Z. Zunic

The paper deals with the conclusions on another new retrospective technique (the volume trap technique) developed at SCK/CEN Mol, Belgium in 1996 year. This technique was field tested in the Balkans (Yugoslavia) also for the first time. A description of the volume trap technique is given in this paper. In simple terms it is the radiochemical measurement procedure of the ratio of the ^{210} Pb / ^{210} Po that accumulates in porous household materials which we call the volume traps (such as sponges retrieved from cushions, chairs, couches, mattresses, etc), due to the decay of radon gas that had diffused into the pores of the material. In principle the measured long-lived radon progeny (^{210} Pb / ^{210} Po) should be directly proportional to the long term average radon in the house and, (unlike the surface trap technique) already described in Paper 1, is independent of aerosol conditions in room air.

As described in Paper I the surface trap retrospective technique was applied successfully in two rural high –radon communities in Yugoslavia , i.e., Kalna and Gornja Stubla. In the areas of Kalna and Gornja Stubla, volume traps were retrieved from about 80 different dwellings, together with data available from surface traps and contemporary radon measurements. (Paridaens et al 2002). As described in this paper the volume trap radon retrospective assessment technique has been applied in over 300 samples, retrieved from dwellings in Serbia, Germany, Norway and Sweden.

All of the 68 non sponge samples taken were a collection of wool, industrial cotton, animal hair, straw and other kinds of house furnishing stuffing. As determined by radiochemical procedures, the overall chemical efficiency was 55% with a standard deviation of 21 %. There is no significant difference between the chemical efficiency for the polyester sponge and the other samples. However, from the volume traps in these areas, it became clear that great care should be taken in the analyses to avoid contamination of the samples with radon daughters, penetrating from the outside into the internal interstitial spaces of the volume traps, for example, as a consequence of dust penetration.

The method presented here for retrospectively assessing radon concentrations has shown to be very robust, whilst comparatively simple in technique. Results obtained show good reproducibility, and in cases where control was possible, they also appeared very reliable. It was concluded that the method will continue to prove its value, especially when applied with other retro techniques, such as the glass surface trap method as already described in Paper 1.

PAPER III - Resume

Integrated natural radiation exposure studies in stable Yugoslav rural communities

The Science of the Total Environment, 2001, Volume 272(1-3), 253-259

Z. S. Zunic, J. P. Mc Laughlin, C. Walsh, A. Birovljev, S. E. Simopoulos, B. Jakupi, V. Gordanic, F. Trotti, R. Falk H. Vanmarckei, J. Paridaensi, K. Fujimoto

In the work described here, on the basis of previous geological and geochemical mapping (Jakupi et al., 1997), attempts were made in Yugoslavia to identify stable rural populations receiving elevated natural radiation exposures which might have potential for future health studies (Zunic et al., 2001a, b). Here the term "stable" is meant to indicate a population having traditionally low inward migration characteristics whose genetic pool has remained relatively stable over many past generations. This makes it more ideal group in which to investigate the health effects of environmental influences than that of a more mobile and genetically diverse urban population. As a result of this approach, two areas were chosen as suitable locations for such a study: (a) the former uranium mining district of Kalna in eastern Serbia; b) Gornja Stubla which is a community in an uraniferous region of Kosovo.

The measurement of indoor radon gas concentrations was carried out using closed passive diffusion alpha track type radon detectors (SSI/NRPB).

As part of the investigation of human exposure to natural radiation in these communities' measurements of penetrating radiation absorbed dose rates in air were made both indoors and outdoors.
In addition to the measurement of radon (^{222}Rn) gas in the dwellings of Gornja Stubla a number of measurements of thoron (^{222}Rn) gas were also made. A special passive alpha track radon/thoron gas detector type of Japanese design supplied by the National Institute of Radiological Sciences, Chiba, Japan, was used for those purposes. This consists essentially of a device with inner and outer measuring chambers, inside each of which there is a plastic alpha track detector. It is designed so that air containing both radon and thoron can rapidly diffuse into the outer chamber while the time of diffusion into the inner chamber is much longer than the half-life of thoron (55 s). Therefore the outer chamber measures both radon and thoron but the inner chamber measures only radon. The difference is measure of the thoron.

In addition to measurements of contemporary radon and thoron concentrations in the dwellings of the field locations a series of ^{210} Po surface and volume trap measurements were also made in the dwellings (Zunic et al. 1999, Paridaens et al. 2000.). The data obtained confirmed the earlier findings described in Papers I and II.

The work described in this paper has identified at least one location (Gornja Stubla) in Yugoslavia where elevated exposures of the general population are being received as a result of exposure to three different natural radiation components (radon, thoron and external penetrating radiation). The combined estimated average annual effective doses from penetrating radiation and radon for these communities, based on the measurements reported here, are 6.8 mSv and 16.2 mSv for Kalna and Gornja Stubla, respectively. When compared to the annual 1 mSv dose limit recommended by ICRP for members of the public these are not insignificant doses for long term exposures.

PAPER IV - Resume

Field experience with volume traps for assessing retrospective radon exposures

The Science of the Total Environment, Vol 272, Iss 1-3, (2001)295-302

Johan Paridaens, Hans Vanmarcke, Zora Zunic and James Mc Laughlin

As already described in Papers I and II (in the case of Volume traps), one looks for sufficiently porous, bulky materials. This is because radon can freely diffuse throughout them and directly deposit its decay products on the internal surfaces of the pores inside the material in the bulk. Here they remain as an indicator of the total radon exposure, until their specific activity is measured by means of a radiochemical separation procedure. The main advantage is that the volume trap directly monitors the radon concentration, so that it is independent of aerosol conditions which greatly influence the accumulation of long lived activity on surfaces in the already described surface trap technique.
The main objective of the study described in this paper was to test this technique in a real field environment.

Seven volume traps collected by the Norwegian Radiation Protection Agency, were received from dwellings in Norway and came from the region of the Hardangerfjord. They were polyester sponges of excellent quality and their ages varied between 16 and 23 years.

Through the European Commission concerted action volume traps were obtained from approximately 12 different dwellings in the Schlema/Schneeberg area. The Schlema/Schneeberg area of East Germany was the location of the extensive uranium mining (now ceased) of the former DDR and has been the subject of major radon epidemiological studies both on miners and members of the public

From the already identified high natural radiation Serbian areas of Gornja Stubla and Kalna, we obtained approximately 140 volume traps of different types.
In addition to these field sample three samples of the porous foam material used in laboratory tests, were placed as new volume traps in carefully monitored dwellings with high radon concentrations in Sweden. The purpose was to test this standard laboratory volume trap material, in field conditions.

It was shown that in some cases care has to be taken in handling the samples, to avoid errors as a result of direct contamination with external radon decay products.

An attempt was made to give an assessment of the achievable accuracy of the method. Where possible, the volume trap retrospective results were compared with contemporary measurements or to retrospective results from surface traps. The overall impression is that although volume traps are sometimes hard to find in the field, the high reliability of the results makes it well worth the effort.

PAPER V - Resume

Correlation between Rn exposure and ^{210}Po activity in Yugoslav rural communities

International Congress Series, Volume 1225 (2002) 87-93

J. Paridaens, Z. S. Zunic, F. Trotti, J. P. Mc Laughlin and H. Vanmarcke

This paper deals with further and continuing studies in high natural radiation areas of Yugoslavia. This study originally started out as an attempt to identify the rural populations exposed to elevated levels of natural radiation. As described in the earlier papers in the communities of Gornja Stubla (Kosovo), Uzice (western Serbia) and Kalna (southeast Serbia), due mainly to geological conditions, the population is exposed to high levels of natural radiation.

In this study, both surface and volume trap retrospective radon assessment techniques were applied.
The results presented in this article concern 33 houses in Kalna, 10 houses in Gornja Stubla and 14 houses in Uzice.

From this field study, it was found that even in stable rural communities, great caution should be taken when using 1-year contemporary radon measurements as representative for a lifetime.
The results obtained clearly show that a carefully planned retro sampling campaign relying on the experience gained during this study would be a considerable asset in any future epidemiological study.

PAPER VI - Resume

High natural radiation exposure in radon spa areas: a detailed field investigation in Niška Banja (Balkan region)

Journal of Environmental Radioactivity, Volume 89 (2006), Issue 3, 249-260

Z. S. Zunic, I. Kobal, J. Vaupotic, K. Kozak, J. Mazur, A. Birovljev, M. Janik, I. Celikovic, P. Ujic, A. Demajo G. Krstic, B. Jakupi, M. Quarto, F. Bochicchio

In the period of 1997-1999, indoor radon surveys were carried out in some regions of Yugoslavia where high radon concentration was suspected. The contemporary and retrospective, both surface and volume trap techniques were applied. Radon concentration was measured in 210 houses by using CR-39 detectors, with a total of 1443 measurements. Detectors were deployed in four seasons in a living room and bedroom of each house, wherever it was feasible. Moreover, retrospective methods were used to measure the activity of radon, long-lived ^{210}Po progeny, on glass and on porous spongy household material. Two high radon-level areas were found at Gornja Stubla (Kosovo) and Kalna (east Serbia), with about 3000 and 500 inhabitants, respectively. The annual effective doses in these areas, estimated according to the ICRP-65 methodology (ICRP, 1993), amounted to 16.2 mSv and 6.8 mSv, respectively.

In 2000, the first indoor radon measurements were conducted in Niška Banja (a spa town located in southern Serbia) using charcoal canisters by the Institute of Health Protection of Workers, Nis, Serbia. Although the measuring sites were not selected on the basis of a defined sampling scheme that took into account geology, hydrology or building material, it appeared clearly that parts of the town were highly exposed to radon. In several rooms, indoor radon concentration was found higher than 10 kBq·m^{-3}. To place this in context it should be noted that the mean indoor radon concentrations in most European countries is less than 100 Bq·m^{-3}.

In order to give a more comprehensive evaluation of exposure and to identify sources of very high radon levels in Niška Banja, the detailed research described in this paper was undertaken in June 2004. Six dwellings with the highest indoor radon concentration were chosen for the measurements. The complementary techniques were applied to determine instantaneous and average radon and thoron concentrations, radon concentration in soil gas, radon exhalation from soil, soil permeability and gamma dose rate. In addition, soil and water samples were collected for further chemical and spectroscopic analyses. The paper describes the results of this detailed investigation in chosen high radon houses in Niška Banja town. From the geological point of view, Niška Banja town is located in the Quaternary alluvium of River Nisava along the contact of Koritnik limestones and thick strata of travertine (spring sediments) also known as ''bigar'' in Serbian literature.

Radon concentrations were found to be very high in soil gas, water and indoor air in Niška Banja town and for the soil gas concentrations have exceeded the upper detection limit (>2 MBq· m^{-3}) of the detector at some places. The concentration of thoron in soil gas was also recorded high (20-46 kBq·m^{-3}).

The measured high radon contents in soil, water and air are due to the presence of high radium contents in the soil and water. However, thorium and potassium contents were found to be within the range of average levels. The high radium contents in the soil and the groundwater may be due to the presence of uranium mineralization in the area. It is also likely that soluble radium bearing material has been transported from

the location of its primary source due to hydro geological leaching and redeposit in the geological strata of Niška Banja. The presence of local faults in the study area may also enhance the radon emanation from the soil.

The results obtained show that the radon concentration in indoor air and soil of Niška Banja can reach extremely high values, correlated with high radium content in soil and high radon exhalation from soil. These results will be utilized to set up the methodology for a more systematic investigation to assess the risk due to high radiation exposure to the general population in Niška Banja town. Guidelines on the use of spa water will also be needed in order to avoid high exposures.

PAPER VII - Resume

Radon survey in the high natural radiation region of Niška Banja, Serbia

Journal of Environmental Radioactivity 92 (2007) 165-174

Z.S. Zunic, I.V. Yarmoshenko, A. Birovljev, F. Bochicchio, M. Quarto, B. Obryk, M. Paszkowski, I. Celikovic, A. Demajo, P. Ujic, M. Budzanowski, P. Olko, J.P. McLaughlin, M.P.R. Waligorski

In this paper the results of a long-term survey of outdoor and indoor radon concentration measurements using Radosys (CR-39) detectors and gamma –ray dose rates in the high natural radiation region of Niška Banja in Serbia are presented. The central parts of Niška Banja town are covered with the formation of the quaternary travertine, a spring sediment formed as a result of carbonate precipitation from spring waters of Niška Banja, which shows a high level of natural radioactivity (Vucic and Pavlovic, 1960). The average Ra-226 concentration measured in soils around Niška Banja is about 900 Bq·m $^{-3}$ (Mazur et al., 2005). Radium –enriched travertine is the main source of radon and, due to its porous structure, permits radon migration, resulting in a high level of radon exposure in the dwellings of this region.

Early in 2000 year, screening indoor radon measurements using charcoal canisters were conducted in Niška Banja by the Institute of Occupational Health Nis (Manic 2006)). Following a request from the local community to confirm those results, in March 2003, a second survey using long-term measurement techniques, by the Institute of Nuclear sciences Vinca, Belgrade, was undertaken.

For the survey 16 houses, with at least two indoor measuring points each, were randomly selected out of 200 previously surveyed measurement points (Manic et al., 2006). In addition, 49 houses not-previously surveyed i.e., the new houses, were selected from the area supposed to be radon prone, giving a total of 65 houses surveyed. The outdoor and indoor radon measurements were conducted from March 2003 to January 2006, during different seasons in order to account for seasonal variations of radon concentrations. A standardized questionnaire was applied and filled in for every house with information provided by the inhabitants. Beside identification information (owner's name, address, GPS coordinates etc.), data about building characteristics were obtained.

Radon concentration measurements were performed using 1 cm^2 CR-39 as an alpha particle detector, enclosed in a small cylindrical diffusion chamber. Tracks on the Cr-39 were counted with an automatic set-up consisting of an optical microscope connected to a CCD camera controlled by a personal computer. Additionally, at most of the indoor and outdoor measurement sites, gamma –ray dose rates (total number 93) were measured using high sensitive MCP-N (LiF: Mg, Cu, P) TL detectors in terms of kerma in air (Budzanowski et al 2004). These rates ranged between 65 and 108 nGy h $^{-1}$.

In this paper are presented results of 80 outdoor measurements and 169 indoor measurements in living rooms and bedrooms of family houses. There were also 95 measurements of gamma dose rate both in indoor and outdoor locations. Outdoor radon measurements were performed during winter, spring and summer seasons in 49 points evenly distributed in the territory of Niška Banja. Outdoor radon concentrations were found to be close to a lognormal distribution considering the season groups separately.

To investigate the different factors affecting radon levels, standard analysis of variance (ANOVA) was applied. Taking into account the lognormal distribution of indoor radon concentrations, natural logarithms of the respective values were considered in this statistical analysis. The dependence of annual

outdoor radon concentration on the type of underground bedrock (travertine and alluvium sediments) was tested and found to be statistically significant.

The correlation between the outdoor gamma dose rates and type of underlying bedrock was tested as well. An interesting result is the significant difference between the standard deviations of outdoor gamma dose rates over travertine and alluvial terrains (test of homogeneity of variances p=0.004). As in the case of outdoor radon a higher average value of outdoor gamma dose rate is associated with the travertine part compared to the alluvium part. The average outdoor radon concentration moving from the alluvium to the travertine part was found to decrease by a factor of 1.6 while the outdoor gamma dose rate decreased by a lower factor (1.3). The higher variability of ^{226}Ra concentration over travertine indicates the higher probability of finding locally elevated radon concentrations both outdoor and indoor.

Measurements of indoor radon concentrations were the main part of the radon survey in Niška Banja. Up to three measurements during winter, spring and summer seasons were performed in the living and bedroom of each dwelling included in the survey. Differences between average values of indoor radon concentrations in dwellings situated over alluvium and travertine parts of Niška Banja were analyzed. The higher average value and standard deviation resulted in doubling the higher percent of dwellings with indoor radon concentrations exceeding the level of 600 $Bq \cdot m^{-3}$ in the travertine part in comparison with the alluvium part of Niška Banja. Higher values of indoor radon concentrations were found to be associated with travertine (p=0.0003).

Following geological and geophysical factors the buildings characteristics are considered to be the second most important factor influencing indoor radon levels. In addition, a quite interesting correlation of indoor radon levels and indoor smoking was obtained. From the analysis of the entire uncategorized data set the highest values were observed in houses of non-smokers while indoor radon concentration was found to decrease with the number of resident smokers. The effect of smokers' presence on indoor radon can probably be associated with higher ventilation rates in homes whose occupants smoke.

The main factor influencing indoor levels in Niška Banja appears to be underlying bedrock type. Highest indoor radon concentrations were obtained for houses located over travertine area compared to the alluvium area. In the group of buildings located in the travertine part and with low natural ventilation indoor radon concentration reaches up to very high levels above 2 $kBq \cdot m^{-3}$. After adjustment for radon seasonal variations the annual indoor radon concentration reached 1630 and 709 $Bq \cdot m^{-3}$ in travertine and alluvium sediments parts of Niška Banja respectively. To place this in context it should be noted that mean indoor radon levels in most European countries are bellow 100 $Bq \cdot m^{-3}$.

Thus the annual effective dose in this case can exceed 50 mSv with a regional average value about 30 mSv. Average outdoor radon levels were found to have relatively high values as well.

According to these observed high values of outdoor and indoor radon the Niška Banja region has to be ranked as a high natural radiation area. The radon survey results strongly indicate that the geological and geophysical factors and the buildings characteristics are primary factors influencing the indoor radon levels. Secondary factors are considered as well.

PAPER VIII - Resume

Comparison of retrospective and contemporary indoor radon measurements in a high-radon area of Serbia

Science of The Total Environment 387 (2007) 269-275

Z.S. Žunić, I.V. Yarmoshenko, K. Kelleher, J. Paridaens, J.P. Mc Laughlin,
I. Čeliković, P. Ujić, A.D. Onischenko, S. Jovanović, A. Demajo, A. Birovljev, F. Bochicchio

For assessing the radon related health risks there is a need for reliable long-term radon data. The two retrospective methods derive the radon concentrations that occurred in dwellings over long periods in the past. These are based on the amount of ^{210}Po which is implanted by alpha recoil into a thin (circa 100 nm) surface layer on glass objects (surface traps ST) or by the amount of ^{210}Po that builds up in the internal air spaces of porous household materials such as spongy filling of furniture, beds etc.(these are volume traps VT).

Due to the high radium content in soil approx 900 Bq·kg^{-1} a number of radon surveys have been conducted both indoor and outdoor and it was found that the average indoor radon concentrations were to be largely dependent upon the type of bedrock on which the dwellings were built (i.e., dwellings built on travertine showed radon concentrations of 1550 Bq·m^{-3} with some dwellings reaching 6 kBq·m^{-3} . For dwellings built on alluvium sediment the average indoor radon concentration was found to be 635 Bq·m^{-3} . Average annual outdoor radon level was found to be 57 Bq·m^{-3} . This is about 6 times the quoted worldwide value by UNSCEAR 2000. A number of retrospective radon measurements (ST and VT) were made to compare with contemporary measurements (CONT). All three (CONT, ST and VT) measurement types were made in the same rooms of the dwellings. Regression analyses made on these measurements showed quite good agreement between the three techniques: For VT-ST r =0.85, for VT-CONT, r = 0.82 and ST-CONT r = 0.73. These results support the agreement between the retrospective and contemporary methods.

PAPER IX - Resume

A campaign of discrete radon concentration measurements in soil of Niška Banja town, Serbia

Radiation Measurements 42 (2007) 1696 - 1702

Z.S. Žunic, K. Kozak, G. Ciotoli, R.C. Ramola, E. Kochowska, P. Ujic, I. Celikovic,
J. Mazur, M. Janik, A. Demajo, A. Birovljev, F. Bochicchio,
I.V.Yarmoshenko, D. Kryeziu, P. Olko

This paper describes the first radon soil gas survey ever carried out in Serbia using passive CR-39 radon detectors implanted in the ground. The radon detectors were buried to a standard depth of 50 cm and laid out over the terrain on a systematic grid pattern. The distance between measurement points was approximately 0.5 km. Measurements were made in a total of 57 locations in the spa town of Niška Banja located in south central Serbia.

In addition to the radon soil gas measurements gamma dose rates in the air were made at a standard height of 1 m above the soil gas measurement points. The gamma dose rate were found to range from 92 to 316 nGyh^{-1} with an average value of 132 nGyh^{-1}

Laboratory gamma analyses, using a NaI(Th) spectrometer to determine natural radionuclides (U, Th and K) in soil samples, were also made. The results of these gamma analyses may be summarized as follows:

^{226}*Ra range: 24-1810 Bq·kg^{-1}, Average was 197 Bq·kg^{-1}*
^{228}*Th range: 11-56 Bq·kg^{-1}, Average was 39 Bq·kg^{-1}*
^{40}*K range: 173-649 Bq·kg^{-1}, Average was 486 Bq·kg^{-1}*

Of these measurements only ^{226}Ra is considered high which made a significant contribution to the gamma dose rate. The elevated ^{226}Ra values were not unexpected as Niška Banja which is a radon spa town with high indoor radon levels. This is also in keeping with the radon concentrations in the soil gas range at a depth of 50 cm. These ranged from 1270 to 155 000 Bq·m^{-3} with an average of 33765 Bq·m^{-3}. As described in the paper it is of interest to note that the soil gas radon followed a bimodal distribution. From this work soil mapping would appear to be a useful predictive technique for indoor radon.

PAPER X - Resume

Radium -226 Concentration in Spring water Sampled in High Radon Regions,

Applied Radiation and Isotopes 2010, 68, 825–827

Aleksandra Onishchenko, Michael Zhukovsky, Nenad Veselinovic, Zora S. Zunic

This paper describes the measurement of ^{226}Ra in spring water in Serbia, Slovakia, Russia and Kyrgyzstan. Based on these measurements, local geology and information on indoor radon levels a main finding of this study is that radium in spring water can be a good indicator of high radium in travertine formations and of radon in dwellings built over such a geological structure. Specifically spring water samples from Niška Banja (N= 55) and from North Caucasus (N=7) were the only ones which showed a high correlation with high radon concentrations in dwellings. In other cases the correlation was weak or non-existent

It was found that travertine ^{226} Radium activity up to 1700 $Bq \cdot kg^{-1}$ where outflow occurred from the springs indicated that there is an increased probability that indoor radon may be high. Therefore while the correlation was not always found to be high measurements of radium concentrations in spring water could usefully serve as an initial feasibility stage of a radon survey in an area.

PAPER XI - Resume

Field Experience on Soil Gas mapping using Japanese Passive Radon/Thoron Discriminative Detectors in Comparison between High and Low Radiation Areas in Serbia (Balkan Region),

Journal of Radiation Research 50 (4), 2009, 355 - 361

Z. S. Zunic , M. Janik, S. Tokonami, N. Veselinovic, I.Yarmoshenko, M. Zhukovsky, T. Ishikawa, R.C. Ramola, G.Ciotoli, K.Kozak, J.Mazur, I. Celikovic, P. Ujic, A.Onischenko, S.K. Sahoo, F. Bochicchio

The work in this paper is concerned with the mapping of radon and thoron concentrations in soil gas in parts of Serbia. It should be noted that the simultaneous measurement of these isotopes in soil gas is an innovative approach. This work was carried out in two different geological regions which had been previously identified as high and low natural radiation areas on the basis of radium in soil measurements.

In both areas passive radon/thoron detectors were implanted in the soil of the sampling points to a depth of 80 cm. For the locations investigated it was noted that the natural radionuclide levels in the soil increased with depth with the values at 80 cm being about 40% greater than those at 40-50 cm.

In Niška Banja soil gas radon values ranged from 1800 to 161000 $Bq \cdot m^{-3}$ with a mean value of 26500 $Bq \cdot m^{-3}$ which is four times higher than in the low background area
Thoron soil gas values in Niška Banja ranged from 900 to 23500 $Bq \cdot m^{-3}$ and with a mean value of 10600 $Bq \cdot m^{-3}$ which is seven times the value in low background area.

From the experience gained in this work it was deduced that proper evaluation of radon and thoron levels in soil gas requires detailed knowledge of physical soil parameters (permeability, diffusion coefficients etc), In order to understand better the dynamical behavior of radon and thoron in soils determinations of these parameters should be carried out in any future work of this type.

6.2 Published papers

1999

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2001

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PAPER IX **Zunic ZS**, Yarmoshenko IV, Kelleher K, Paridaens J, McLaughlin JP, Čeliković I, Ujić P, Onischenko AD, Jovanović S, Demajo A, Birovljev A. Bochicchio F. Comparison of retrospective and contemporary indoor radon measurements in a high-radon area of Serbia. Science of The Total Environment, Volume 387, Issues 1-3, 15 November 2007, 269-275

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2010

PAPER X Aleksandra Onishchenko, Michael Zhukovsky, Nenad Veselinovic, **Zora S. Zunic.** Radium-226 Concentration in Spring Water Sampled in High Radon Regions. Applied Radiation and Isotopes, 2010, 825–827

PAPER I

Radiation Measurements

PERGAMON

Radiation Measurements 31 (1999) 343–346

THE USE OF SSNTDS IN THE RETROSPECTIVE ASSESSMENT OF RADON EXPOSURE IN HIGH RADON RURAL COMMUNITIES IN YUGOSLAVIA

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ABSTRACT

A description is given of the field application of a technique using CR-39 and LR 115 detectors to determine alpha recoil implanted ^{210}Po surface activity on domestic glass artefacts in dwellings. These investigations took place in two small stable rural communities in uraniferous areas of Yugoslavia where between 32% and 74% of contemporary indoor radon levels were found to be above the commonly used Action Level of 200 Bq m^{-3} and individual levels as high as 8700 Bq m^{-3} were measured. The ^{210}Po data is used to retrospectively estimate radon exposures in these communities. Comparisons between the retrospectively estimated radon exposures and those being received at present are made.

KEYWORDS

Retrospective radon; ^{210}Po in glass.

INTRODUCTION

A number of major case-control epidemiological studies on lung cancer incidence in the general population exposed to radon in their homes have taken place in recent years or are in progress (Lubin and Boice, 1997; Darby *et al.*, 1998). In most of these studies contemporary radon measurements made in the present dwellings of the study subjects have been used as surrogates for radon concentrations in past decades even though changes in radon levels and residence may have occurred. In this paper an account is given of the principles and some field applications of a retrospective technique, using the alpha track detectors CR-39 and LR 115, to measure ^{210}Po implanted in glass surfaces (surface traps) by means of which the cumulative radon exposure in a dwelling in past decades may be estimated.

POLONIUM-210 SURFACE TRAPS

Short-lived radon progeny may deposit on available surfaces in dwellings thus giving rise over time to a build up of long-lived progeny. On surfaces such as glass activities of ^{210}Po may arise as a result of the decay of recoil implanted activity following the alpha decay of surface deposited ^{218}Po or ^{214}Po. Surface activities of ^{210}Po on glass surfaces in dwellings generally range from a few to some hundreds of Bq m^{-2} for extremely high radon exposures. Under typical room living conditions, a ^{210}Po surface activity of about 1 Bq m^{-2} might be expected for a radon exposure of 1 kBq m^{-3} for 1 year. The measurement of such alpha recoil implanted ^{210}Po surface activity on glass or other vitreous materials in dwellings can be used to make a retrospective assessment of radon exposure (Samuelsson, 1988). Based on room models describing the behaviour of radon and its progeny in rooms, models have been developed for the purpose of estimating the mean radon concentration in the past responsible for a

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measured ^{210}Po activity on a room surface (P[58] ndörfer *et al.*, 1978; Falk *et al.*,1996). Thes[58] dels are sensitive to the values of a number of key room parameters. These principally are the characteristics of the room aerosols, the room surface to volume ratio and the ventilation rate. In a large-scale survey of dwellings the range of these parameters in the past cannot be known and even their present values can only be estimated. In this work by means of a questionnaire on house characteristics and occupant habits each of the key room parameters was estimated to lie in either the low, medium or high category of the values covering the likely ranges of the key parameters in normal dwellings (Knutson, 1988). The values of the key parameters chosen on the basis of the completed questionnaire and the age of the selected glass artefacts are used in the model to estimate the radon concentration in the past. The uncertainties involved in this approach are recognised but by using the likely full range of room parameters, it is possible to estimate the range of values within which the mean long-term radon level would lie.

A recently developed method for large-scale fieldwork, the "CR-LR difference technique", using alpha track detectors was used in the work described here to measure ^{210}Po (Falk *et al.*, 1996). In this technique, a few square centimetres of each of the alpha track detectors CR-39 (polyallydiglycol carbonate) and LR 115 (cellulose nitrate) are mounted side by side on a chosen flat glass surface. For standard etching conditions the LR 115 is sensitive to alpha particles in an energy window in the approximate range 1.2 - 4.8 MeV while the CR-39 is sensitive to alphas with energies from less than 1 MeV to much greater than the maximum energy (7.68 MeV) emitted by radon progeny. As the ^{210}Po alpha energy is 5.3 MeV the LR 115 will not record tracks from the surface trapped ^{210}Po but will produce tracks proportional to the intrinsic alpha activity naturally present in the glass. The CR-39 track density will thus be a mixture of tracks due to the implanted ^{210}Po alpha emissions and those from the intrinsic activity of the glass. The following expression gives the surface ^{210}Po activity for a glass surface onto which side by side CR-39 and LR 115 detectors were mounted for T hours:

$$P = (CR - B \times LR)/(T \times K) \qquad (1)$$

where P = ^{210}Po activity in Bq m^{-2}, CR = CR-39 net tracks per cm^2, LR =LR 115 net tracks per cm^2, B = CR-39 : LR 115 intrinsic alpha activity track density ratio for unexposed glass and K= PIC (Pulse Ionisation Chamber) determined CR-39 sensitivity factor to ^{210}Po. B and K values depend on the actual etching regimes and track acceptance criteria used. For standard procedures values of B = 1.97 and K = 0.081 tracks cm^{-2} per Bq m^{-2} hr were obtained (Falk *et al.*, 1996). In carrying out field retrospective radon exposure investigations using CR-LR difference technique strict protocols were used to select suitable glass artefacts and for the handling of the detectors. Primarily because of the likely occurrence of anomalous radon progeny deposition rates window glass and glass surfaces over heaters, in draughts etc are to be avoided where possible. The age and exposure history of the glass chosen should be well known. The glass in the frame of the picture of an important family event, such as a wedding, often fulfils these requirements. Such a glass may be seen as a quasi-personal radon dosimeter as it usually will remain with a family even when there is a change of residence. The typical exposure period used in this work for the alpha track detectors mounted on glass was about three to four months. The CR-39 alpha track densities recorded for these exposures were mainly within the range 5 to 25 tracks mm^{-2}. Measured ^{210}Po values ranged from about 3 to 30 Bq m^{-2} with individual values as high as 50 Bq m^{-2}. Contemporary indoor air radon gas concentrations were measured in the chosen dwellings by means of standard passive diffusion type CR-39 radon detectors with a calibrated radon sensitivity of 2.6 alpha tracks cm^{-2} kBq^{-1} m^3 hr.

FIELD WORK

Fieldwork was carried out in two small rural communities in Yugoslavia. These are Kalna in southeast Serbia and Gornja Stubla in Kosovo. In both these areas, elevated concentrations of uranium and thorium are present in rocks and soil. In Kalna some uranium mining has taken place in the past. A recent investigation of radon concentrations in soil gas and indoors in Kosovo indicated Gornja Stubla to be a high indoor radon community with individual concentrations as high as approximately 2500 Bq m^{-3} being detected (Jakupi *et al.*, 1997). In the present work, 8700 Bq m^{-3} was the highest value detected. In this community many of the houses are built substantially from local rock which has a

uranium content of the order of 10^{-5} g/g (~125 Bq kg^{-1}). Corroborative evidence of elevated natural radioactivity was obtained in the present work by HP Ge gamma spectrometry of soil from Gornja Stubla which yielded the following mean values for natural radio-nuclides: ^{238}U = 392 Bq kg^{-1} ; ^{226}Ra = 249 Bq kg^{-1} ; ^{232}Th = 181 Bq kg^{-1} and ^{40}K = 1751 Bq kg^{-1}. In addition, elevated penetrating radiation dose rates as high as 400 nGy hr^{-1} were detected in some dwellings.

In the present work as indoor radon concentrations often have a strong seasonal variability a sequential series of four radon gas measurements, each typically of 3 months duration, were carried out in most of the 106 dwellings chosen. This yielded an annual mean radon value for each dwelling. These results are summarised in Table 1. For comparison purposes, the UK and Finland are presented as examples of countries having low and high mean radon levels respectively while Ireland represents the more typical European situation (ECA, 1995).

Table 1. Indoor Radon Gas Concentrations

Community or Country	No. of Dwellings Surveyed	Radon conc. (Geom. Mean) [Bq m^{-3}]	Geom. Std. Dev [Bq m^{-3}]	Percent over 200 Bq m^{-3}
Gornja Stubla	34	361	2.3	74
Kalna	72	151	1.8	32
U.K	2093	15	2.2	0.5
Ireland	1259	34	2.5	3.8
Finland	3074	84	2.1	12.3

Most countries have been found to have regions with indoor radon levels substantially above the national mean and it is clear that Kalna and Gornja Stubla are in this elevated radon category. An estimate of the likely contemporary annual effective dose to the dwelling occupants may be obtained by using a current ICRP conversion factor of 1 mSv per 60 Bq m^{-3} (ICRP, 1993). In 64 of the houses in both these areas it proved possible to choose suitable glass artefacts for the in-situ measurement of surface ^{210}Po activities in order to make an assessment of radon exposures in past decades. Glass artefacts chosen were in the age range 7 to 70 years which covers well the period of about three decades which is considered to be the approximate latency period for radon induced lung cancer. The measured contemporary and the model estimated retrospective radon concentrations in the dwellings investigated are shown in Figure 1.

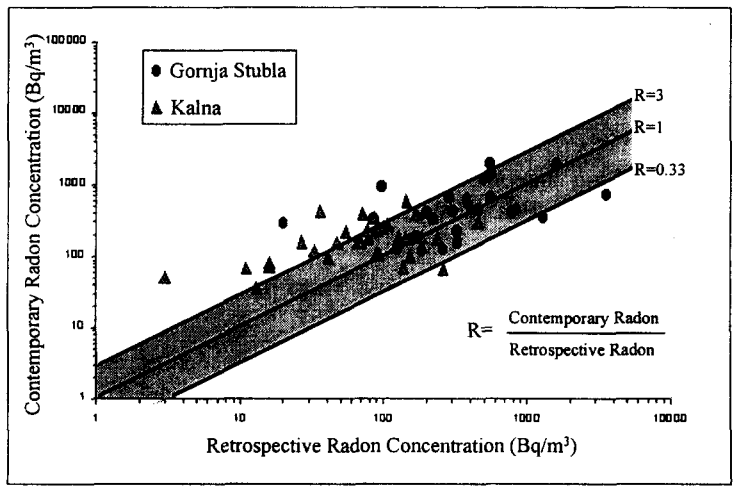

Fig. 1. A comparison between estimated retrospective and contemporary radon concentrations.

Defining the quantity R as being the ratio between the contemporary and estimated retrospective radon concentrations the value for each dwelling is presented in Fig. 1 together with a band of R values centred about the line R=1 (i.e. no change in radon values between past and present). Due to the uncertainties involved in estimating room parameters, it is considered reasonable to expect that the estimated value of R for a dwelling may lie within a factor of at least three of its true value. As the majority of points in Figure 1 lie within the band from R=0.33 to R=3 this is taken as a good indication that there has been no significant change in mean radon levels in these two communities over recent past decades. It should be noted that long term radon stability appears strongest for the Gornja Stubla data. Additional studies are in progress, involving other European and Japanese colleagues, on radon, thoron and penetrating radiation investigations in these communities.

CONCLUSIONS

On the basis of the results obtained to date it is clear that Kalna and Gornja Stubla are elevated radon communities and the estimated retrospective radon concentrations indicate that radon exposures in these dwellings in the recent past decades have not changed significantly. These areas of Yugoslavia may therefore be considered as suitable potential radon epidemiological study locations.

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PAPER II

PERGAMON

Applied Radiation and Isotopes 53 (2000) 361–364

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Retrospective radon assessment by means of ^{210}Po activity measurements

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Abstract

A method for retrospectively assessing the average radon concentration in a dwelling, for a period as long as 40 years or more, is described. It is based on the free penetration of radon gas into bulky spongy materials called volume traps, and subsequent trapping of radon decay products inside them. This leads to a build-up of ^{210}Pb, a radon decay product with a half life of 22.3 years, which reaches an equilibrium with the alpha emitter ^{210}Po, indicative of the average radon concentration over the exposure period. The stuffing of mattresses, chairs, cushions, etc. can be considered a good volume trap. A chemical separation procedure is described, transforming the volume trap into a ^{210}Po alpha source, whose activity is determined through alpha spectroscopy. The technique has been applied in over 300 samples, retrieved from dwellings in Serbia, Germany, Norway and Sweden. The technique has proven to be very robust and reproducible. In cases where radon data were available during the exposure period of the volume trap, a remarkable agreement was found. The best volume traps appeared to be polyester sponges found in many stuffings. The major drawback is the destructive nature of the method, its major advantage is the direct relationship between the radon gas and the ^{210}Po signal. The technique can be useful in epidemiological studies of the lung cancer risk from indoor radon, for assessing the long term radon exposure.

Keywords: Radon; ^{210}Po; Exposure assessment

1. Introduction

When trying to assess the public health detriment, caused by radon exposure in the living environment, through epidemiologic studies amongst the general public, the problem of determining the radon exposure of the subjects with sufficient accuracy immediately arises. A common practice is to use current, often short term, radon concentration averages, and to simply extrapolate these to time periods as long as 40 years. This can lead to very large errors in the estimate of the radon exposure, thus compromising the epidemiological results. Radon concentrations are known to vary a lot, and relatively common interventions such as changing the windows of a house, can drastically change the current radon values with respect to the past. Moreover, there are well-known examples of dwellings, built directly on former mining shafts, where the radon concentration has varied over orders of magnitude, depending on the mining conditions. Also,

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dwellings constructed on glacial deposits can show very fluctuating radon concentrations, depending on atmospheric conditions. Therefore, it is advisable to rely on retrospective methods, for assessing long term radon concentrations. These methods are based on the build-up of low levels of ^{210}Pb and, consequently, ^{210}Po activity, that can be related to the long term average radon concentration.

2. Method

Basically two methods of retrospective radon assessment exist. One is a surface trap technique, in which airborne radon decay products deposit on smooth glass surfaces. Subsequently, they can be implanted through recoil, and remain fixed in the near surface layer of the glass, where they can be detected through alpha decay by means of track etch detectors or pulse ionisation chambers (Samuelsson, 1988). The other is a volume trap technique (Oberstedt and Vanmarcke, 1996). In this case, it is the radon gas itself which diffuses freely into bulky, porous materials, depositing there its decay products. The short lived decay products are trapped in the volume trap, and give rise to a build-up of ^{210}Pb. After about 1.5 year, equilibrium is reached between ^{210}Pb and ^{210}Po, which is an alpha emitter with a half life of 138 days. It is this isotope that has to be separated for analysis. Therefore, a small sample of the material is taken from the centre of the volume trap. It is important that the sample could not have been contaminated with airborne decay products during its lifetime, hence, the need for avoiding the surface of the volume trap. The volume of the sample has to be determined. This can be done by weighing if the density is known, or less precisely by cutting the sample in a shape that fits into a recipient of known volume. In practice, for polyether volume traps, samples with a weight between 0.5 and 3 g are usually taken, representing volumes between 15 and 100 cm^3. The sample is then put into a round bottom flask, together with an accurately known volume of a calibrated tracer solution. This tracer solution was commercially bought, and contained 99.6% of ^{208}Po, 0.37% of ^{209}Po and 0.03% of ^{210}Po according to the manufacturing firm. Periodically, the total alpha activity of this solution is checked by direct deposition on a stainless steel plate, and counting in a ZnS counter. In the first stage, the sample has to be brought in solution, after which the solution is evaporated, leaving just the dry residual. Therefore, first 100 ml of concentrated HNO_3 solution is added to the sample. The recipient is attached to a water cooled reflux cooler. The solution with the sample is then boiled for 2 h, with the purpose of destroying the matrix of the volume trap material. Now, the solution is poured into a clean beaker, and the glass recipient is rinsed twice with 25 ml of concentrated HCl solution, and this is added to the beaker. The beaker is placed under an infrared lamp, until the liquid is evaporated. Now, 5 ml concentrated HCl is added to the beaker, and then evaporated again. This procedure is repeated until no brown fumes escape from the solution anymore and a dry residual remains in the beaker.

In the second stage, the auto-deposition of the ^{210}Po on a silver plate takes place. Therefore, the dry residual is redissolved in 2 ml of concentrated HCl, and this is poured together with some 20 ml of distilled water into a plastic bottle. This procedure is repeated once more, this time with 40 ml of distilled water, in order to realise the complete transfer from the beaker into the plastic bottle. Distilled water is added, to obtain about 100 ml of solution, and then 100 mg of ascorbic acid is added. On top of the plastic bottle, a degreased silver cupel, supported by a stainless steel one, is placed and the tap is screwed on top of the bottle to seal it. The plastic bottle is now turned upside down, and a few needle holes are made in the bottom of the bottle for the excess pressure to escape. The bottle is placed in a heated spot (50–70°C) and stirred every now and then. Ideally, it should be placed on a vibrating surface, to allow continuous homogenisation of the solution. The auto-deposition of the polonium isotopes now begins and is allowed to continue for at least 48 h. After the auto-deposition, the cupel is retrieved and rinsed with a little alcohol. The alpha activity is determined by means of alpha spectrometry, allowing to separate the three lines present in the spectrum, ^{208}Po (5.115 MeV), ^{209}Po (4.880 MeV), ^{210}Po (5.3044 MeV), without any problem. The chemical efficiency can be determined from the tracer isotopes and is typically between 30 and 70%. Once the total ^{210}Po activity that was present in the sample is known, this has to be related to the original radon concentration. It is straightforward to calculate the amount of radon atoms that have decayed inside the volume trap, giving rise to this ^{210}Po activity, at a certain time, as long as the exposure period is known with sufficient accuracy. However, to convert this to radon gas concentration, we must know what the radon absorption capacity of the original volume trap was. In some cases, this just depends on the volume fraction of air present in the volume trap, but in other cases, extra radon adsorption in the volume trap material is possible. It is wise to investigate this for each separate type of volume trap material (Paridaens and Vanmarcke, 1999).

3. Results

In Fig. 1, the chemical efficiency is shown for a total of 325 samples. From these, 257 were polyester

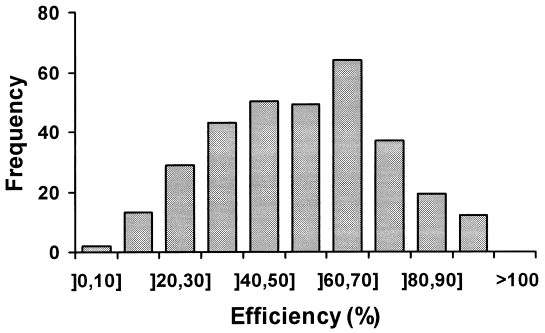

Fig. 1. Chemical efficiency, obtained with 325 volume trap samples. The average chemical efficiency was 55%, with a standard deviation of 21%.

sponges, retrieved from cushions, chairs, couches, mattresses, etc. primarily from the rural regions of Kalna and Gornja Stubla, in Serbia. There were also 18 Norwegian samples and 28 from the Schneeberg, former mining area, in Germany. The 68 non sponge samples were a collection of wool, industrial cotton, hair, straw and other kinds of stuffing. The overall chemical efficiency was 55% with a standard deviation of 21%. There is no significant difference between the chemical efficiency for the polyester sponge and the other samples. In fact, the sponges separately yielded an average of 52% and a 19% standard deviation, whereas the rest yielded 64% with a standard deviation, of 27%. It is very probable that the chemical efficiency could still be improved, by letting the autodeposition to take place on a vibrating surface, unfortunately, no such equipment was available. With one type of volume trap material, a problem was encountered. It concerned a natural latex mattress, which appeared to be not dissolvable in nitric acid in the first stage of the procedure. Here, probably solvents like toluene should be tried in the first stage.

A laboratory test was performed to test the reliability of the method for very low radon exposures. Therefore, 20 samples were taken from a batch of polyester sponges, that were exposed to an integrated radon concentration equivalent to an exposure to 62.3 $\pm$ 4.2 Bq/m^3 during 20 years. Lower radon concentrations would not be deemed very important in epidemiological studies. An ingrowth of 70% of the equilibrium concentration of ^{210}Po was awaited for. Ten of the samples had a volume 23 cm^3, the others were 78 cm^3. These are the two typical sample sizes that we use for analysing volume traps retrieved from dwellings. Table 1 shows the results obtained from these analyses. First, we notice that for both sample sizes, the results from the analyses do not differ significantly from the exposure value and the standard deviation does not exceed 20%, for either of the used sizes. These results were obtained with acceptable alpha counting times of about 6 days for the smaller and about 4 days for the larger sized samples, resulting in 87 counts and 123 counts under the ^{210}Po peak, respectively. The average chemical efficiency was seen to be larger for the smaller sized samples: 64% as opposed to 41%. This indicates that an optimum should be looked for in the analysed sample size, combining a high ^{210}Po count rate, with reasonable amounts of solvents used in the procedure.

Table 1
Polyester sponges exposed to an equivalent radon concentration of 62.3 $\pm$ 4.2 Bq/m^3 during 20 years[a]

Volume (cm^3)	23	78
Found radon concentration (Bq/m^3)	59.3	58.1
Standard deviation (Bq/m^3)	11.1	9.9
^{210}Po counts per day	14	29
Total of ^{210}Po counts	87	123

[a] Results in the columns are averages over the 10 samples for each of the volumes.

Three fresh polyester sponges were exposed in three different rooms of a Swedish dwelling for 2.25 years. The radon concentration was monitored during the whole of the exposure time and the average radon concentrations in the three rooms were 2500, 2090 and 2360 Bq/m^3, respectively. The values obtained from the sponges were 2500, 2000 and 1900 Bq/m^3, which is very satisfactory indeed.

In the areas of Kalna and Gornja Stubla, volume traps were retrieved from about 80 different dwellings. From the same dwellings, data will be available from surface traps and contemporary radon measurements. This will be completed with thoron measurements, and gamma dose measurements, and the idea is to obtain a complete picture of the various exposures to ionising radiation for the population of these areas (Zunic et al., 1999). Comparison between all these different data sources is still very incomplete, but seems to further indicate the reliability of the volume trap method. For example, there were 22 cases where two volume traps taken from the same dwelling were available. In 16 of those cases, the time period covered by the two volume traps was the same. The location of the two volume traps in the dwelling, however, was mostly very different. For each dwelling, the difference between the two obtained retro exposures was expressed as a percentage of the average of the two. If we average this percentage over all 22 cases, we find 34% and a median of 34%; if we only consider 16 cases covering the same time period, we get an average of 28% and a median of 19%. Considering the large variability of radon concentrations in general and the fact that the two volume

traps were not even necessarily in the same room, this is a remarkable agreement. However, from the volume traps in these areas, it became clear that great care should be taken in the analyses to avoid contamination of the samples with radon daughters, penetrating from the outside into the heart of the volume traps, for example, as a consequence of dust penetration (Paridaens et al., 1999).

4. Conclusion

The method presented here for retrospectively assessing radon concentrations has shown to be very robust, whilst comparatively simple in technique. The results obtained show good reproducibility, and in cases where control was possible, they also appeared very reliable. The major sources of error are not in the method itself but in the knowledge of the volume traps exposure period, the age of the volume traps, and the sometimes tricky business of determining the volume of arbitrarily shaped objects. The major advantage is the direct relationship between the actual radon gas concentration and the measured signal. A drawback is the destructive nature of the method, making it hard sometimes to find suitable volume traps in dwellings. The method has, however, been successfully applied on a large scale in the areas of Kalna and Gornja Stubla, helping in the construction of a total radiation exposure picture for the general population there. We think the method will continue to prove its value, especially when applied with other retro techniques, such as the glass surface method.

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PAPER III

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Integrated natural radiation exposure studies in stable Yugoslav rural communities

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Abstract

The results of field investigations of natural radiation exposures of the general population in two stable rural communities in Yugoslavia are presented. The principal emphasis was on exposures to contemporary indoor radon, but measurements of external penetrating radiation absorbed dose rates in air were carried out in the majority of cases. In addition, in a limited number of dwellings, measurements of thoron gas concentrations were made. By means of making a series of sequential 3-month radon measurements, both seasonal variations and annual average radon levels in the dwellings were determined. Using passive alpha track detectors, individual radon and thoron indoor concentrations as high as 9591 Bq m^{-3} and 709 Bq m^{-3}, respectively, were detected while absorbed dose rates in air in the dwellings as high as 430 nGy h^{-1} were recorded. On the basis of these different types of measurements, assessments could be made of the integrated natural radiation exposures being received by the populations. In addition to contemporary radon measurements, retrospective radon exposure assessments in most of the dwellings were made on the basis of measurements of ^{210}Po concentrations in both surface (glass) traps and in

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volume (porous materials) traps. A description is given of the sampling strategies and protocols used in this field work. It is shown that at least one stable rural community receiving high natural radiation exposures, has been clearly identified and plans for future health investigations of the population there are outlined.

Keywords: Natural radiation; Radon; Thoron; Yugoslavia

1. Introduction

Most of the evidence of the detrimental health effects of exposure to ionizing radiation has been obtained from studies of cohorts or populations which have received high doses and/or high dose rates. A central conservative tenet of radiation protection philosophy is the linear hypothesis in which it is assumed that the risk is linearly proportional to the dose with no threshold. Direct evidence of the validity of the linear hypothesis at low doses (i.e. a few mSv $year^{-1}$) is, however, difficult to obtain. The findings of recent case-control epidemiological studies of radon associated lung cancer in subjects drawn from the general population in a number of countries may help to address this issue. There are, however, many difficulties associated with such studies due to the effect of confounding factors such as smoking, the low exposure of subjects and subject residence mobility (McLaughlin, 1999). At the level of the individual subject a high mobility makes it difficult to assess long-term radon exposure and it also means that the population being studied does not share a common stable genetic pool over a long period of time. In the work described here, on the basis of previous geological and geochemical mapping (Jakupi et al., 1997), attempts were made in Yugoslavia to identify stable populations receiving elevated natural radiation exposures which might have potential for future health studies (Zunic et al., 1997a,b). As a result of this approach, two areas were chosen as suitable locations for such a study: (a) the former uranium mining district of Kalna in eastern Serbia; (b) Gornja Stubla which is a community in an uraniferous region of Kosovo in south-eastern Yugoslavia.

2. Measurement of indoor radon concentrations

The measurement of indoor radon gas concentrations was carried out using closed passive diffusion alpha track type radon detectors. The particular type used was the SSI/NRPB design in which the alpha track registration medium is CR-39 and the housing shell is black conducting plastic. In order to further reduce possible effects on the detector response due to static electricity each CR-39 foil was dipped in an antistatic solution before each detector was assembled. The detector calibration factor is a function of the etching and track counting procedures. For the participating laboratories the radon calibration factors were in the range of 2–3 tracks cm^{-2} kBq m^{-3} h. These calibration factors were determined by participation in a number of NRPB (UK) radon intercomparison exercises. Track counting was made by means of both automatic image analysis and manual optical microscopy.

In order to obtain a realistic value of the mean annual indoor radon concentration in the chosen dwellings a sequential series of detectors were placed in one or more living areas over a twelve

Table 1
Summary statistics for radon gas measurements

Community	Number of dwellings surveyed	Number of detectors deployed	Percentage of detectors recovered
Kalna	75	391	98.7
Gornja Stubla	65	616	89

month period. Each single detector remained exposed in a dwelling for a period of approximately 3 months and was then immediately replaced by a new detector. In this way a total of 4 such detectors were placed at each chosen location thus yielding both season and annual average radon concentration values. In a small number of dwellings when a detector could not be recovered at the end of an exposure period, due to absence of occupant, etc., the unrecovered detector was allowed to remain for a further 3 months until finally it was recovered. The summary statistics for the radon gas measurements are shown in Table 1.

The results of the indoor radon gas measurements in the dwellings of Kalna and Gornja Stubla are shown in Fig. 1a,b. Also shown are the estimated annual effective doses being received by the occupants of these dwellings. Although there are many radon exposure to annual effective dose conversion factors (DCF) available in the literature here we used the recently reported DCF of 7.3 mSv WLM^{-1} (Porstendorfer and Reineking, 1999) which is considered to be more applicable to aerosol conditions in normal dwellings than the approximate 4.2 mSv WLM^{-1} DCF as used by the ICRP for high aerosol conditions (ICRP, 1993). For exposure at home the ICRP assuming 7000 h $year^{-1}$ indoors and an equilibrium factor of 0.4 uses a coefficient for annual exposure per unit radon concentration of 4.40×10^{-3} WLM per (Bq m^{-3}). Combining this with the DCF chosen here translates in terms of annual radon exposure to approximately 31 Bq m^{-3} year mSv^{-1}. The doses in Fig. 1a,b are calculated on this basis. It is fully realized by the authors that such conversions cannot be taken as applicable at the level of an individual but applied to a group they are of assistance in assessing the likely ranges of effective doses being received. When compared to world and European average values it is obvious from Fig. 1 that both the radon levels and estimated doses are anomalously high in both these communities and in Gornja Stubla in particular. At the individual house level some strong ($> 25\%$) seasonal variations of radon concentrations were observed with winter values being greater than those in summer. When radon data were aggregated and averaged over each community the seasonal variation was approximately 5%.

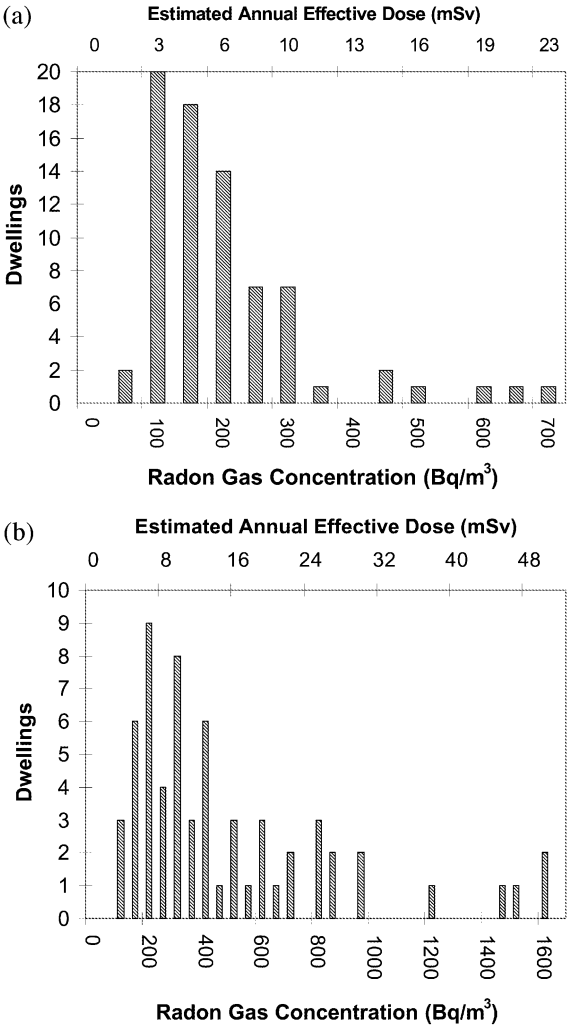

Fig. 1. Indoor radon concentrations and estimated annual effective doses. Mean annual radon gas concentration: (a) Kalna (75 dwellings), (b) Gornja Stubla (62 dwellings).

3. Penetrating radiation measurements and doses

As part of the investigation of integrated human exposure to natural radiation in these communities measurements of penetrating radiation absorbed dose rates in air were made both indoors and outdoors. Previous geological and

geochemical studies in Gornja Stubla had indicated that above normal penetrating radiation levels might be expected due to the elevated uranium and thorium content of the local rocks and their extensive local use as building materials (Jakupi et al., 1997; Gordanic et al., 1999). The absorbed dose rates in air were measured using a hand held Environmental Radiation Meter Type 6-80, Mini-Instruments (UK). The sensor is an energy compensated G-M tube modified to reduce the non-linear energy response which is characteristic of G-M tubes. This instrument type was calibrated over the approximate energy range 0.5–1.3 MeV at the Berkeley Nuclear Laboratories (UK). While the instrument response is not ideal it was considered adequate for absorbed dose rate in air measurements under field conditions. Conversion from absorbed dose in air (Gy)

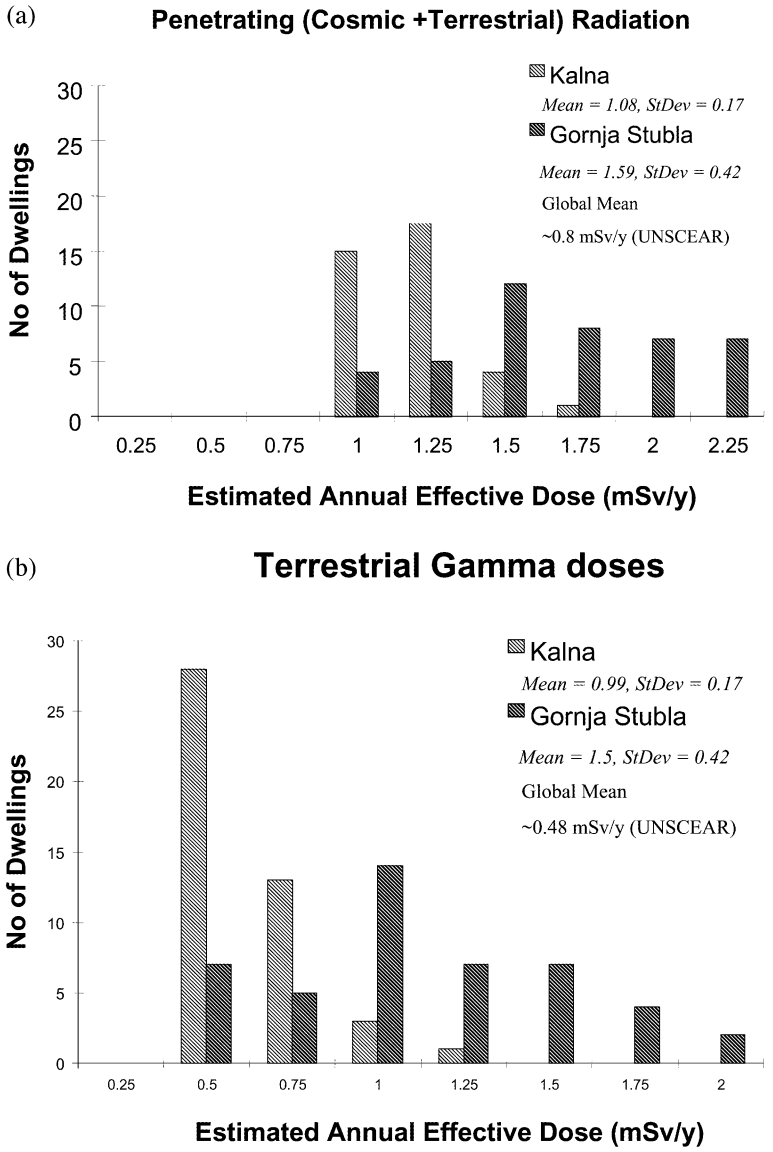

Fig. 2. Estimated annual effective doses from penetrating radiation: (a) total, (b) terrestrial.

to effective dose (Sv) was made using a conversion factor of 0.7 Gy Sv^{-1} which is recommended for the general population but a higher conversion factor for children might be more applicable (UNSCEAR, 1993). The measured total absorbed dose in air includes the instrument response to cosmic radiation. The cosmic radiation response was experimentally determined by measuring the instrument response from a small boat in the middle of the Danube river in Belgrade at a distance of approximately 425 m from the nearest land at which distance the terrestrial radiation contribution to detector response was negligible. In this manner the cosmic radiation response was found to be equivalent to approximately 0.06 microGy h^{-1} or 0.36 mSv $year^{-1}$. This estimated cosmic radiation dose rate is very similar to literature quoted population weighted northern hemisphere average values. In Fig. 2a,b the estimated total (gamma + cosmic) annual doses and those from terrestrial radiation alone for the two communities are respectively presented. In calculating annual effective doses the UNSCEAR approach was adopted in which an indoor occupancy factor of 0.8 was assumed.

As in the case of radon exposures the penetrating radiation doses in these two communities are well above world averages ($\sim$ 0.8 mSv $year^{-1}$ for total penetrating and 0.48 mSv $year^{-1}$ for terrestrial radiation) in particular in the case of Gornja Stubla.

4. Thoron gas measurements

In addition to the measurement of radon (^{222}Rn) gas in the dwellings of Gornja Stubla a number of measurements of thoron (^{220}Rn) gas were also made. A special passive alpha track radon/thoron gas detector type was used for this purposes. This Japanese developed detector consists essentially of two interlocking and interconnected plastic hemispheres (Doi et al., 1992). Air containing both radon and thoron diffuse into the first hemisphere where alpha tracks from both gases and their alpha emitting progeny are recorded on the surface of an alpha track detector. The air exchange rate between the two hemispheres is much smaller than the decay constant of thoron ($T_{1/2} = 55$ s) so that the alpha track signal recorded by the alpha track detector surface within the second hemisphere is essentially due only to the radon series. By means of calibration these track densities and the difference between them allows separate measurements of the radon and thoron concentrations to be made.

Using these detectors measurements of thoron and radon were made in a total of 44 locations in 23 dwellings in Gornja Stubla. Because of the short half-life of thoron the detectors were placed within 15 cm of walls which are the assumed source of the thoron. The thoron results are presented in Fig. 3. Thoron concentrations as high as 750 Bq m^{-3} were recorded. It should, however, be pointed out that in some cases very large uncertainties are associated with these thoron values. Nevertheless, these thoron measurements clearly indicate that elevated exposures to thoron progeny are likely to be taking place in Gornja Stubla dwellings. No measurements of thoron progeny were made, but they may be made in the future. As the double hemispherical detector units measure both radon and thoron it is interesting to compare the measured concentrations of both these gases at each measuring point. This comparison is shown in Fig. 4. The correlation coefficient was found to be 0.63 at the 0.05 significance level. Even though there are large uncertainties associated with each data point this degree of correlation may be due to the fact that the local building material is rich in both the thorium and uranium series although in the case of radon the main source is probably the subjacent ground (Gordanic et al., 1999). Further work is obviously needed to identify the principal sources in these dwellings for both gases.

While thoron levels in these dwellings are high, and reflect the thorium content of the local geology and building materials, in the absence of information on thoron progeny data it is impossible to make any estimate of doses from the thoron series. In fact, even if thoron progeny data were available it is clear that great difficulties exist in making meaningful dose estimates (Nuccetelli and Bochicchio, 1998).

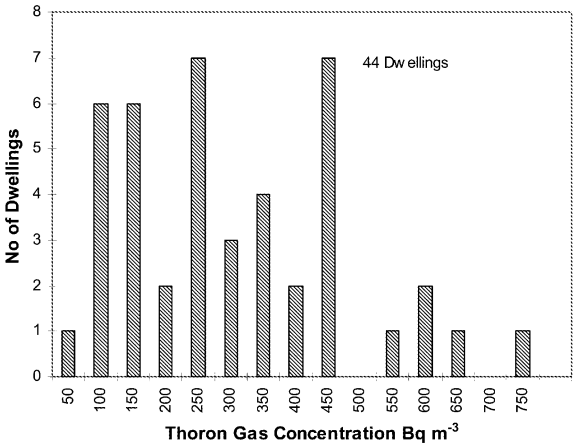

Fig. 3. Indoor thoron concentrations in 44 dwellings of Gornja Stubla.

5. Long lived radon progeny measurements

In addition to measurements of contemporary radon and thoron concentrations in the dwellings of the field locations a series of ^{210}Po surface and volume trap measurements were made (Zunic et al., 1998, 1999). In the case of surface traps the technique used was by means of alpha track detectors mounted on suitable glass objects and by analysing the resulting alpha tracks to determine the surface activity of ^{210}Po that had arisen as a result of short-lived radon progeny plateout and subsequent recoil implantation. Different surface trap alpha track detector techniques from laboratories in Sweden, Ireland Norway and Italy (the so-called 'SINI' intercomparison exercise) were used in this work. In the volume trap technique ^{210}Po within the interstitial spaces of samples of porous sponge-like material taken from the dwellings was determined radiochemically at the Belgian Nuclear Research Centre (SCK/CEN), Mol. Each of these two ^{210}Po trap techniques make it possible to retrospectively estimate the radon concentrations in the dwellings in past decades. As in the case of the contemporary radon and thoron measurements all of the field work was carried out by the Institute of Nuclear Sciences, Vinca, Belgrade. The results and findings of both the SINI exercise and the volume trap work are reported elsewhere (Zunic et al., 1999) and in this volume (Paridaens et al., 1999).

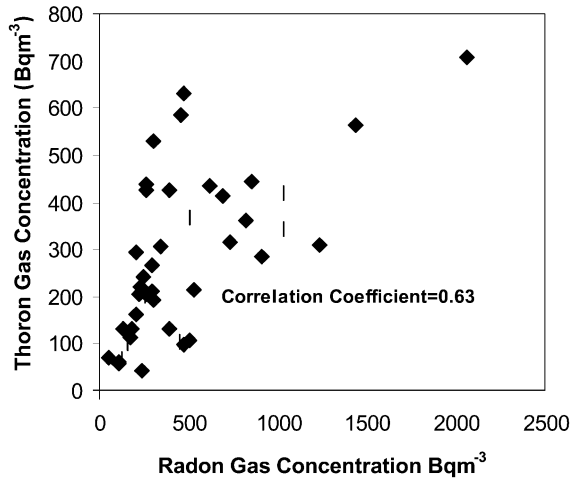

Fig. 4. Correlation of thoron and radon concentrations in 44 dwellings in Gornja Stubla.

6. Conclusions and future work

The work described in this paper has identified at least one location (Gornja Stubla) in Yugoslavia where elevated exposures of the general population are being received from three different natural radiation components (radon, thoron and external penetrating radiation). The combined estimated average annual effective doses from penetrating radiation and radon for these communities, based on the measurements reported here, are 6.8 mSv and 16.2 mSv for Kalna and Gornja Stubla, respectively. Because of the stable genetic nature of this rural community and the absence of industrial pollution this community can be considered as an almost ideal natural laboratory to investigate the health effects of natural radiation. It is likely, because very similar geological and geochemical characteristics extend over many kilometers around Gornja Stubla, that other nearby rural communities may also have elevated natural radiation exposures. It is therefore intended that further similar field work will take place in order to better delineate the extent of this elevated natural radiation zone. In parallel with this it is intended to initiate studies in the

general population in this zone and also in Kalna to ascertain if there are any health effects associated with the elevated radiation exposures. Studies of this type have already taken place in some high natural radiation regions of the world, most notably in Kerala (India) and Yangjiang (China) (Steinhausler, 1993). The results of these studies have been somewhat equivocal for a variety reasons mainly due to such factors as the presence of confounding factors, statistical uncertainties of national cancer data and deficiencies in the design and implementation of these studies. There were, however, some indications (such as increased frequency of chromosomal aberrations in circulating blood lymphocytes) that were statistically associated with the above normal radiation exposures but a causal relationship was not established. In the planned health studies in these Yugoslav communities a careful study design will be made but before its implementation it will be sent for critical review to independent experts.

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PAPER IV

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Field experience with volume traps for assessing retrospective radon exposures

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Abstract

Approximately 200 volume traps were retrieved from dwellings in various radon prone areas in Europe. They were analysed for the purpose of retrospective radon assessment. Emphasis is put on specific problems encountered when using field samples as opposed to laboratory exposed samples. It was seen that in very dusty circumstances, direct penetration of radon decay products from the outside to the centre of the volume traps calls for extra caution. Rinsing the samples is proposed as a solution and was tested in field and laboratory conditions, showing good results. An attempt was made to give an assessment of the achievable accuracy of the method. Where possible, the volume trap retrospective results were compared with contemporary measurements or to retrospective results from surface traps. The overall impression is that although volume traps are sometimes hard to find in the field, the high reliability of the results makes it well worth the effort.

Keywords: Exposure; Radiation; Radon; Monitors; Radiation; ^{210}Po

1. Introduction

Retrospective assessment of radon exposure (RARE), is a technique which has developed greatly in recent years. The ultimate aim is to provide reliable radon exposure data as a support to epidemiological studies concerning the health effects of radon exposure in the living environment, amongst the general public. Therefore, the ^{210}Po activity is measured in objects originating from dwellings of the investigated subjects. (Lively and Ney, 1993; Samuellson and Johansson, 1994) In the case of volume traps, one looks for sufficiently porous, bulky materials. Radon can freely diffuse throughout them and directly deposit its decay products in the bulk. Here they remain as an indicator of the total radon exposure, until they are set free by means of a radiochemical

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separation procedure, destroying the actual sample. The main advantage is that the volume trap directly monitors the radon concentration, so that it is independent of aerosol conditions, which are influenced by complex processes such as turbulence or air movement. This direct dependence on radon gas concentration, eliminates the need of using elaborate room models for obtaining the retro results. The feasibility of the technique has been demonstrated in earlier laboratory tests (Oberstedt and Vanmarcke, 1996), using our 'standard' polyester sponges as volume traps. The same type of material can often be found in the padding of cushions or in chairs or mattresses. The main objective was to test this technique in a real field environment. We wanted to see if it was possible to find suitable materials on the field, and to determine criteria by which to prefer certain materials over others or to reject certain materials. Moreover, we wanted to develop specific procedures for dealing with different types of volume traps in order to obtain the best possible results.

2. Materials and retrieving areas

In principle, any bulky, sufficiently porous material, with known age and history, can serve as a volume trap. It suffices that radon gas can diffuse freely through the material on a time scale short compared with the radon half-life, so that the radon concentration inside the volume trap is continuously representative for the radon concentration outside the volume trap. The material should, however, be such that radon decay products cannot migrate in to or out of the volume trap. Moreover, the volume trap material should not contain a significant natural ^{210}Po background, so as to disturb the measurements. Results are based on the detection of the ^{210}Po isotope, through alpha spectroscopy on a source produced by chemically destroying the volume trap material and plating out the ^{210}Po activity (Oberstedt and Vanmarcke, 1996). An obvious, easy to find candidate, seemed to be wood. Although wood complies with the first two conditions, it was shown (Paridaens and Vanmarcke, 1999) that the high and variable ^{210}Po background renders wood unsuitable as a volume trap. Another important factor in choosing a suitable material for a volume trap is the ease with which its volume can be determined. We must be able to determine the volume it actually occupied in situ during its life as a volume trap. The retrospective radon value directly depends on this volume, since it determines the volume of air sampled by the volume trap and hence the amount of ^{210}Po to be expected inside for a certain radon concentration. This is all but trivial if you think of certain materials such as industrial wool or cotton, used in stuffing of cushions, etc. Errors of 100% or more in volume estimation are easily imaginable for these materials. In practice, the best available material remains the polyester type of sponge one often finds in mattresses, sofas, cushions and so on. Here also it must be stressed that sometimes this material, especially in cushions, is found, cut in very small pieces. This again poses the problem of assessing the volume occupied by the material in situ. Moreover, in this case, one can never be sure that the material was not recycled from objects used before in other places, thus rendering the history of the volume trap uncertain. The destructive character of the technique, makes it difficult to convince people to donate volume traps: people do not get their volume traps back. All of this brings us to what certainly is the major drawback of the technique: in some cases it is very difficult to obtain suitable items. However, this notwithstanding, we were able to retrieve a very significant number of volume traps from several interesting areas:

- Seven volume traps were received from dwellings in Norway and came from the region of the Hardangerfjord. They were polyester sponges of excellent quality and their ages varied between 16 and 23 years.
- We were involved in the EC concerted action: retrospectively estimated radon in areas affected by uranium mining activities. Through this collaboration we obtained volume traps from approximately 12 different dwellings in the Schlema area.
- From the Serbian areas of Gornja Stubla and

Kalna, we received approximately 140 volume traps of different types.

Three samples of the material we used in laboratory tests, were placed as new volume traps in carefully monitored dwellings with high radon concentrations in Sweden. They were collected after approximately 27 months, and for each sample two separate analyses were made. The purpose was to test our standard volume trap material, in field conditions.

2.1. Experimental methods

Determining the volume of the sample is often the most critical part of the analysis. When the density of the sample is known, this can of course be obtained by simple weighing, but only when we use our standard polyester material. Mostly, we simply have to cut the samples so that they fit into a receptacle of known volume. We routinely use a 23 cm^3 or a 78 cm^3 shape, depending on the amount of volume trap material available. As a statistical test, for each of the two shapes, we cut 10 pieces of our standard polyester material to fit into them. By weighing we obtained the exact volume of each of these pieces, which gave us an idea of the accuracy of our method for determination of volume.

For each of the Norwegian samples two or three separate analyses were performed. From these dwellings data were available from contemporary radon measurements and from retro glass surface samples. Contemporary radon concentration measurements here show a variation of as much as three orders of magnitude, within sometimes 24 h (from approx. 50 Bq/m^3 up to over 50 000 Bq/m^3). This makes obtaining an average radon concentration from contemporary values practically impossible, and thus shows the stringent need for integrating retrospective techniques. No useful comparison was possible with contemporary measurements. Comparison could be made between different analyses of the same samples, and where available with retro glass samples.

The Schlema area is a well known area in the east of Germany, where mining activities used to take place. The different stages of these mining activities have caused the radon concentrations to vary considerably during the last decades. It was difficult to collect volume trap samples in this area, since it appeared that lots of people recently had all of their furniture renewed, throwing out the older objects in the process. However, some polyester sponges, wool or cotton stuffing from cushions, and some plumes from cushions could be retrieved. Ages varied between as much as 60 years and only a few years. Retro measurements on glass samples and data from contemporary radon were also available for these dwellings.

Gornja Stubla and Kalna are geologically very stable, rural areas, with a stable population. Most of the samples retrieved there were between 15 and 20 years old. Approximately 70% of them were polyester sponges from mattresses, chairs, cushions, etc. of very different sizes, shapes, densities and degree of cleanliness. The rest of the samples were mostly industrial wool or cotton stuffing, or even straw or seaweed. From these dwellings data were available from contemporary radon measurements and from retro glass surface samples, allowing comparison between different methods. Very often we obtained more than one volume trap from the same dwelling, allowing interesting comparisons. For a lot of cases, the samples obtained here contained an excessive amount of dust which had penetrated to the very centre of the volume traps. It was soon seen that this could cause problems as will be discussed later. As a solution, rinsing the samples under tap water, hence removing all dust, was proposed. The effect of this rinsing was investigated by comparison between rinsed and non rinsed pieces from the same field volume trap. Moreover, a laboratory experiment was set up where of 20 samples who had received the same radon exposure, half were thoroughly rinsed and half not, and results were compared.

Finally, to investigate the uncertainty on the obtained results, close to limit of the usability of the technique, a low exposure radon exposure equivalent to 62.3 ± 4.2 Bq/m^3 during 20 years was given to our standard polyester material in a radon chamber. From this material 10 pieces of 23 cm^3 and 10 pieces of 78 cm^3 were cut and

analysed. It gave an idea of the spread of the obtained results, as a function of the volume used for analysis.

For all analyses, when pieces of the same volume trap of unknown density were analysed, an attempt was made to asses the density of the sample as accurately as possible. If its shape was regular, this was done by assessing the volume of the whole volume trap through measurement, and subsequent weighing. Otherwise, pieces were cut to fit a known volume and then weighed, and density was obtained as an average. In all cases, the volumes of the analysed pieces of the same volume trap were then recalculated using their individual masses and common density.

3. Results

For the 10 volume assessments, using the 23 cm^3 shape, we found an average $\mu = 23.0$ cm^3, with a standard deviation $\sigma = 1.4$ cm^3 or $\sigma_{\%} = 6.1$ %. With the 78 cm^3 shape we found $\mu = 77.8$ cm^3, $\sigma = 2.7$ cm^3 or $\sigma_{\%} = 3.5$ %. The histogram in Fig. 1 shows the spread of the results obtained from the 20 samples, 10 large and 10 small ones, exposed to the same integrated radon concentration of 62.3 ± 4.2 $Bq/m^3 \times 20$ years. We find $\mu = 58.7$ and $\sigma = 9.9$ $Bq/m^3 \times 20$ years or $\sigma_{\%} = 16.9$ %. If we look at the small (23 cm^3) and large (78 cm^3) samples separately, we would find averages of $\mu_s = 59.3$; $\sigma_S = 11.1$; $\sigma_{S\%} = 18.7$ % and $\mu_L = 58.1$ $\sigma_L = 9.0$; $\sigma_{L\%} = 15.5$ % $Bq/m^3 \times 20$ years, respectively. This means that there is no significant difference in the accuracy that can be obtained using an over three times larger volume for analysis. It must be stressed that the values of σ_S and σ_L are not determined by errors in volume determination itself. All of the volumes were derived through exact weighing, knowing the material density, rendering errors in these volumes negligible. Moreover, part of the difference between σ_S and σ_L can be explained by simple counting statistics. For the larger volumes an average of 123 counts were collected during on average 362 902 s, or approximately 4.2 days, i.e. 29 counts per day. For the smaller we had 87 counts in 523 660 seconds or approximately 6.1 days, i.e. 14 counts per day. The efficiency of the chemical separation seems to drop with larger samples, but this will be subject of further investigation. Allowing the same counting statistics for the smaller samples obviously becomes a problem because it takes too long. It is one of the major limiting factors for the technique. If we combine the two sources of error, volume determination, and analyses process itself, we would find: 20% S.D. for the smaller samples and 16% standard deviation for the larger ones. It goes without saying that this is excellent indeed, but we must stress that it does of course not take specifics of the volume trap characteristics into account. Was the volume trap taken from a suitable place, do we know its age correctly, was it always in the place we think it was, was it impermeable to direct contamination with radon decay products, did it retain all radon decay products deposited inside, etc.? These are all questions we must be able to answer to assess the reliability of the result. A 25% error in age estimation for example is easily possible, thus contributing more to the error in the result than the whole analysis procedure. However, these problems are common to all retrospective techniques, and in practice, field results are usually fairly good as we will see now.

Table 1 shows the results for the Norwegian volume traps. Where available, the retro glass result is added. We always noticed a good agreement between different pieces of the same volume

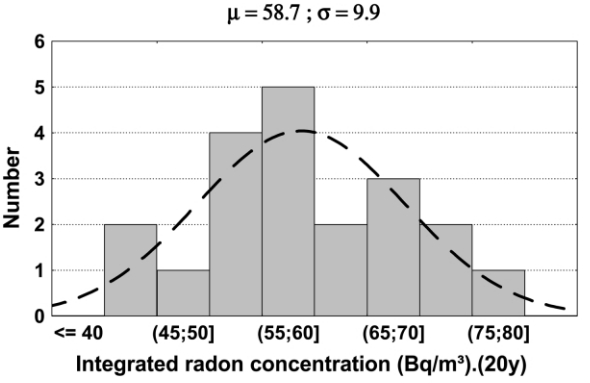

Fig. 1. Result of the analyses of 20 pieces of volume trap material, all taken out of a batch that received an integrated 62.3 ± 4.2 $Bq/m^3 \times 20$ years radon exposure. Half of the pieces had a volume of 23 cm^3, the other half 78 cm^3.

Table 1
Results from the Norwegian volume traps

Sample name	Sample age (years)	Retro radon concentration (kBq/m^3)	Weighted sample average (kBq/m^3)	Retro glass result (kBq/m^3)
NW6	20	1.5		
		1.5	1.5	0.5
NW13	16	8.3		
		11.5		
		10.5	9.3	11.0
NW17	26	1.0		
		1.1	1.1	
NW18	25	0.13		
		0.090	0.11	
NW20	17	3.2		
		3.3	3.2	3.3
NW21	20	6.1		
		6.7	6.6	
NW22	23	0.58		
		0.82		
		0.55	0.61	0.38

trap, fully in line with what can be expected from the standard deviations obtained from the laboratory tests. Comparison with the retro glass results is very good for NW13 and NW20, fairly good for NW22, and rather bad for NW6. However, it must be said that mostly the glass and volume trap samples do not cover the same period of time, because they are different objects. Taken into account the uncertainties of both methods, these results are very satisfactory. As soon as available, comparison with the missing retro glass results will be made.

Table 2 shows the results for the standard volume trap material placed in a dwelling in Sweden. Once again all samples were analysed twice, showing excellent agreement between them. Contemporary radon concentrations were available over the period of exposure of the volume traps. Although the situation is rather complex since it concerns measurements in different rooms of a house over a rather long period, there seemed to be a good agreement between contemporary and retro results.

Table 2
Results from the standard volume trap material placed in a Swedish dwelling

Sample name	Sample age (years)	Retro radon concentration (kBq/m^3)	Weighted sample average (kBq/m^3)	Contemporary radon (kBq/m^3)
VRUM	2.25	2.7		
		2.3	2.5	2.6
MRUM	2.25	1.9		
		1.9	1.9	2.1
SUVRUM	2.25	2.3		
		1.9	2.0	2.4

For Schlema, from the point of view of volume traps, a few interesting results can be quoted. They are summarized in Table 3. Also here, the polyester sponge volume traps were analysed twice each, yielding once more excellent agreement. The overall impression from comparison with other results is very satisfactory, especially of course where the other results actually came from the same room and from a comparable exposure period. Differences of a factor two in results need not necessarily be bad, seen the complexity of the involved techniques. It is seen that even in the case of wool samples, especially for the 1512DP sample, nice agreement is obtained between all results. In the case of the feathers, the probable cause of largest error is the volume assessment. It is very hard to assess the volume of a pack of

Table 3
Some interesting results from the Schlema area

Code/ type	Age (years)	Retro C_{Rn} (kBq/m^3)	Weighted average	Other results for comparison (kBq/m^3)
1112EP/ sponge	35	0.21 0.24	0.22	0.054, 0.057, 0.146; other room, comparable ages
3711DP/ sponge	35	4.1 4.2	4.11	1.1, 3.46, 3.5; same room, probably older ages
3801AQ/ sponge	20	0.70 0.71	0.71	1.3, 0.94, 0.40, 0.471, 0.53; same room, different ages
3801DR/ sponge	30	0.72 0.72	0.72	1.1, 0.81, same room, different ages
1512DP/ wool	25	0.13	0.13	0.042, 0.068, 0.101, 0.080, 0.107; same room, different ages
1511DP/ wool	17	6.6	6.6	3.2, 12.0, 3.7; different room, much older samples
4002AP/ feathers	60	0.87	0.87	0.25, 0.13, 0.15, 0.20; same room, different ages

feathers occupied during its lifetime inside a cushion.

An overwhelming amount of volume traps reached us from the areas of Gornja Stubla and Kalna. It would be impossible and probably useless to quote all the results here. Rather, we shall focus on some striking findings in these areas. Due to the rather primitive nature of the dwellings in this area, some of the volume trap samples contained a very high content of dust. The dust had penetrated sometimes 10 cm deep, down to the very centre of the samples. Since a lot of these dirty volume traps, yielded very high results as compared with the cleaner ones, suspicion arose, that radon decay products had been able to penetrate from the outside into the volume trap, along with the dust. To check this, some volume traps were sliced into pieces from the (dirty) edge to the (sometimes cleaner) centre and further to the opposite (dirty) edge. Fig. 2 shows two such profiles, one for a sponge with very dirty edges and a very clean centre, the other for a sponge that was dirty throughout. It can be seen that large errors are possible even when little dust is present in the samples, as in slices 2 and 4 of SK1. This inhomogeneous behaviour of spongy volume traps was only seen in the presence of clearly visible dust contamination. As a solution, rinsing under running tap water was proposed, to try and remove the dust and along with it the 'extra' radon decay products. It was seen in cases where we disposed of both a clean and a dirty sample of the same dwelling, that rinsing the dirty sample yielded results comparable with results from the clean one. It was also seen that for samples with a clean centre and a dirty edge, that rinsing the dirty edge brought the results very close to those of the clean centre. This is illustrated in Table 4. Moreover, we had many cases where we had two different dirty samples from the same dwelling. Analysing them before rinsing often gave very different results, after rinsing, results were always much closer to one another. All of this seemed to indicate that rinsing under

Table 4
The effect of rinsing dirty spongy samples

Sample	Remarks	Retro C_{Rn} (kBq/m^3)
B3	Clean central	0.31
	Dirty edge	1.6
	Dirty edge, rinsed	0.32
IN1	Dirty, central	0.94
	Dirty, central, rinsed	0.15
IN1 – II	Other sample from same dwelling, clean, central	0.13

tap water might be a good procedure to deal with those dirty sponges. To check if this rinsing procedure did not eliminate the ^{210}Po activity deposited by the radon gas, call it the right activity, we took 20 samples out of a batch of standard polyester material, all exposed to a radon concentration equivalent to 62.3 ± 4.2 Bq/m^3 during 20 years. Ten of them were analysed as such, 10 were very thoroughly rinsed under running tap water before analysis. We obtained for the non-rinsed: $\mu = 57.2$; $\sigma = 8.7$ and for the rinsed: $\mu_r = 60.2$; $\sigma_r = 11.2$. It shows that no significant difference was induced by the rinsing. A lot of spongy volume traps from the areas of Gornja Stubla and Kalna where analysed more then once, and the results always confirmed the good agreement, very similar to Tables 1–3. A source for concern still remains the volume traps consisting of industrial wool or cotton we received from this area. They persistently yielded higher results than the spongy volume traps from this area, as can be seen in Fig. 3. Such behaviour was not seen in the Schlema area, where volume trap samples contained little or no dust. It is therefore presumed that this problem too might be caused by direct penetration of radon decay products, however, due to the physical nature of these samples, rinsing here seems much more difficult. Those materials will be the subject of further investigation.

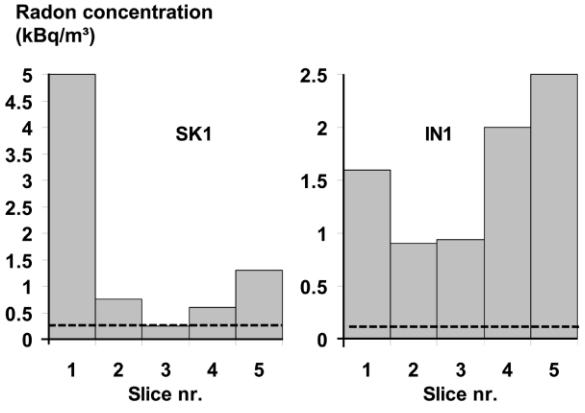

Fig. 2. Two profiles: SK1 was a sponge with very dirty edges and a very clean centre, IN1 was a sponge that was dirty throughout. All slices were 2 cm thick. The thick dashed horizontal lines show the correct result. We can see that only the sponge with the clean centre produces the correct result, and only when the sample is taken sufficiently centred.

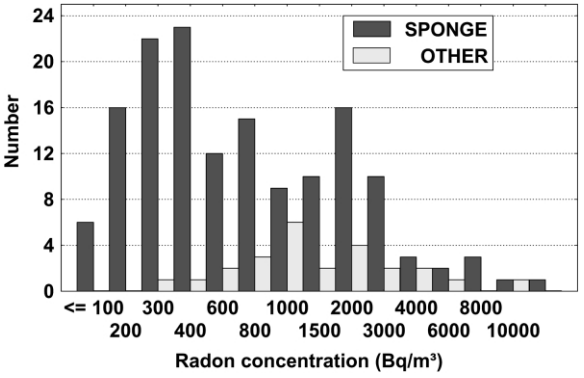

Fig. 3. For the areas of Gornja Stubla and Kalna, it is seen that the results obtained with polyester sponges are on average lower than those obtained with other volume traps such as industrial wool or cotton. This phenomenon was not observed in other areas.

4. Conclusion

Intrinsically, the volume trap technique, is a very powerful and robust technique for assessing retrospective radon concentrations. The volume trap directly monitors the radon concentration and the resulting independence of elaborate room models is a large advantage. This was now also seen in the field. As far as the sample history data are reliable, a precision of within 30% seems easily achievable, even for low radon concentrations of less then 100 Bq/m^3 during approximately 10–20 years. Results are very consistent for multiple analyses of the same sample or for analyses of multiple samples from the same dwelling. It was shown that in some cases care has to be taken in handling the samples, to avoid errors as a result of direct contamination with external radon decay products. More specifically, for very dusty samples, rinsing before analysis is recommended. The problems of sample collecting do not seem insurmountable, since only very small amounts of material are needed to perform a reliable analysis. Polyester sponges are by far the best choice of material, industrial cotton or wool

may also be used, although some further investigation of the behaviour of these materials will still be necessary.

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PAPER V

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Correlation between Rn exposure and ^{210}Po activity in Yugoslavian rural communities

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Abstract

Background: In epidemiological studies investigating radon-related health risk, the radon exposures are often derived from contemporary short-term radon measurements. Recent radon measurements, taken over a period of 1 year in dwellings, are often casually extrapolated and taken to be representative for periods spanning several decades. By simply considering the well-known variability of radon concentrations in dwellings and its sensibility to trivial factors such as living habits or minor changes in the dwelling itself, an amount of skepticism concerning the reliability of this practice should arise. In mining areas, this variability is already well acknowledged since mining conditions, such as ventilation of mine shafts, flooding of mine galleries, etc., are known to be able to change radon concentrations in dwellings built on top of mine shafts for over more than one decade. *Methods*: This study was conducted in stable rural communities in Yugoslavia. Present radon concentrations were compared to an indicator of past radon concentrations, that is the ^{210}Po activity, fixed on glass surfaces or in the bulk of voluminous materials. *Results*: In less than 50% of the cases, the past and present radon concentrations agree within a factor of 2. *Conclusions*: This comparison indicates that even in stable rural communities, deriving past radon concentrations by extrapolating present ones is not a particularly good practice.

Keywords: Epidemiology; Retrospective radon assessment; Dose reconstruction

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1. Introduction

This study originally started out as an attempt to identify the rural populations exposed to elevated levels of natural radiation [1,2]. In the communities of Gornja Stubla (Kosovo), Uzice (western Yugoslavia) and Kalna (southeast Yugoslavia), due to geological conditions, the population is exposed to high levels of natural radiation. These populations might be considered to form a potential cohort for future health studies. Therefore, gamma, radon and thoron measurements were carried out in these regions. It was soon realised that radon exposure was the largest contributor to the radiation dose and that information on past radon concentrations was essential.

2. Methods

In recent years, basically two methods for assessing past radon concentrations have been developed. These retrospective methods are both based on the trapping of ^{210}Pb, a radon decay product with a half-life of 22 years. Its accumulation is related to the integrated radon concentration over periods that can be as long as 80 years, long enough to be of interest for epidemiology. The ^{210}Pb eventually decays into ^{210}Po, an easily detectable alpha emitter. This ^{210}Po activity is used to assess long-term radon exposures. However, although both methods are based on the same measurable parameter, they fundamentally differ. The first, called the surface trap (ST) method, looks at smooth surfaces, mostly glass. Airborne radon decay products can be deposited and implanted through alpha recoil into the glass surfaces. This results in the ^{210}Po alpha activity on glass surfaces [3–5]. The second, called the volume trap (VT) method, looks at the bulk of porous bulky materials, mostly polyester spongy stuffings of chairs, mattresses, etc. The radon gas can freely diffuse into these materials and decay inside them, thus depositing its decay products there. Airborne radon decay products ideally should not be able to penetrate the volume trap. This again results in the ^{210}Po activity inside the bulk of the volume trap [6–8]. The most important difference between the two methods is that the ST method continuously registers the amount of airborne radon decay products and not directly the radon concentration, while the VT method continuously registers the radon concentration in the air but not the airborne radon decay products. The VT technique is destructive and cannot be done in situ. The ST technique is not destructive and can be mostly performed in situ. There is something to be said for both methods. The radiation dose depends on the airborne radon decay products rather than on the actual radon concentration. The ST method is more likely to yield information on this. On the other hand, the radon concentration can be more directly calculated from the VT method using simple volumetric considerations rather than a room model. Combining both seems to be the ideal situation and can be indicated in future studies. Both methods can suffer from the conditions of excessive dust or dirt. In the VT technique, this can lead to dust penetrating into the bulk of VT, and this dust contains extra radon decay products, leading to higher radon estimates. The opposite can happen with the ST technique since the presence of layers of dust on the glass can partly prohibit the implantation of radon decay products, hence leading to lower radon estimates.

In this study, both techniques were applied. The ST technique, at that time still at an experimental stage, was applied for the first time on a large scale in actual field conditions. Later on, the VT technique that was developed somewhat later and was therefore even more at an experimental stage was added where it was possible. This campaign is one of the first large field tests for both methods, certainly as far as VT was concerned. As both methods matured, interesting results became available [1], strongly indicating the need for a retrospective component in studies like this one.

3. Results

The results presented in this article concern 33 houses in Kalna, 10 houses in Gornja Stubla and 14 houses in Uzice. They were selected on the basis of the fact that for most of them, results were available from 12-month contemporary (CONT) radon measurements, one or more volume traps, and one or more surface traps. Some of the ST results were derived from an earlier Swedish–Italian–Norwegian–Irish (SINI) intercomparison exercise in Uzice and Gornja Stubla [9]. The rest of them were produced by the UCD in Dublin. All VT results were produced at the SCK-CEN in Mol. Fig. 1 shows a distribution of the age of VT and ST. It is striking that the average age of ST is almost twice that of VT. About 25% of ST are over 45 years old, while there is no VT of this age. This was rather to be expected: it is easier to find glass objects, such as mirrors, picture glass or indoor window panes, that have survived such long periods than polyester stuffings. The latter tends to wear within 10–20 years, after which they are replaced. Also, in rural communities, older stuffings are more likely to consist of wool, straw, cotton, etc. We must also say that usually, the exact age of glass items is easier to determine than in the case of VT. People might not remember exactly when they bought their current sofa or mattress, but they do

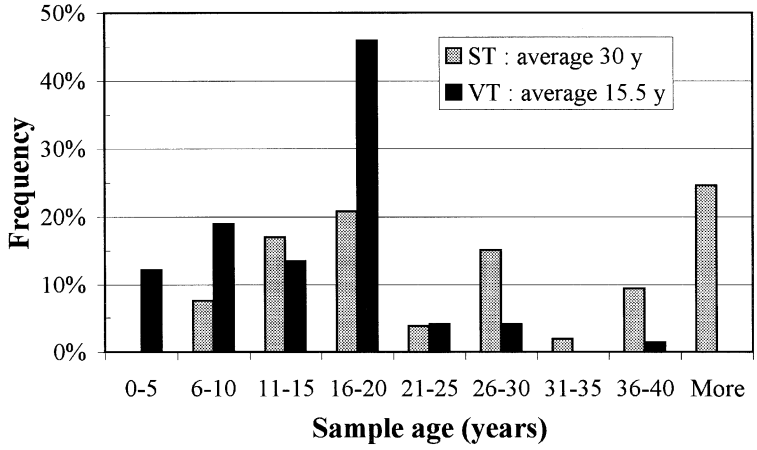

Fig. 1. Distribution of the ages of volume traps (VT) and surface traps (ST).

remember when their house (with glass) was constructed or when they got married (picture glass). It was interesting to see that out of 74 VT samples, 26 of them or 35% were stated to be exactly 20 years old. Moreover, 8% were said to be exactly 15 years old and 11% exactly 10 years old. This gives us 54% of the VT samples with ages equaling exactly the round numbers 10, 15 or 20 years, indicating that these ages must be seen more as an indication rather than as exact values. Of course, the age of the sample directly influences the estimated long-term radon concentration, and therefore, is an important source of possible error. The fact that ST on the average has been exposed twice as long as VT also contributes to the possible differences in radon concentrations derived for the same house from different techniques. Therefore, it is interesting to look at the Uzice subgroup first. Here, the average ages of ST and VT are 16.6 and 17.6 years, respectively, almost equal. Radon concentrations in this region are fairly low. The average CONT is 110 Bq/m^3 with a maximum of 291 Bq/m^3 and a minimum of 32 Bq/m^3. Fig. 2 shows, on a logarithmic scale, the ratio of the retrospective radon estimate to the CONT. The horizontal dashed lines indicate the zones where this ratio is between 0.5 and 2. This means a difference of not more than a factor of 2 between the retrospective and CONT. For 8 out of 14 houses, the VT result lies within this zone. For ST, this is 6 out of 13. In only the first three out of 13 possible cases where both VT and ST are within a factor of 2 of CONT, that is only 23%. None of the ratios exceed a factor of 5, and for most of the houses, both VT and ST results point in the same direction. Exceptions to this are house numbers 14 and 10. For number 14, VT indicates that during the last 10 years, the radon concentrations were on the average five times higher than now, whereas ST suggests that during the last 20 years, the radon concentrations were on the average twice as low as that of the present. This is of course impossible, and it is a clear case where further investigation is needed to obtain a reliable radon estimate for this house. No explanation is available at present. Either of the retro samples might have been faulty and the CONT radon measurement might be as well.

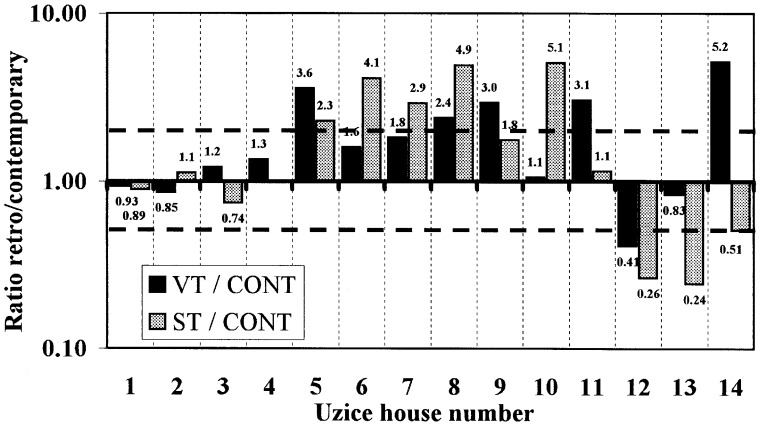

Fig. 2. Ratio of VT and ST retrospective radon estimates to contemporary radon concentrations in Uzice. The dashed lines indicate the zone where retro and contemporary results are within a factor of 2 of one another, that is a ratio of >0.5 and <2.

Possibly, works may have been performed in this house, drastically altering the radon concentrations, or the items might simply have been retrieved from different rooms in the house, while CONT is at the average value for the whole house. Whatever the reason is, the CONT value should not be blindly taken as representative for longer periods or for the whole house. A similar conclusion can be drawn for house 10. For houses 5_9, and perhaps also 11, both VT and ST seem to indicate that the past radon concentrations here were a factor of 2–4 higher in the past than today. The opposite goes for houses 12 and 13. Only houses 1_4 show fairly constant radon concentrations over long periods.

In the Kalna region, the contemporary radon average is 190 Bq/m^3 and it varies between 29 and 457 Bq/m^3. All samples collected here were relatively old, on the average, 17.5 years for VT and 39.5 years for ST. There is however a large difference in the average age between both types of samples. Fig. 3 shows that 17 VT results and 10 ST results fall within a factor of 2 of the CONT result, that is 57% and 37% of the available houses, respectively. In only the first six of the 24 houses, where both retro techniques were available, both retro results fall within a factor of 2 of CONT, that is only 25%, comparable with the Uzice case. Out of the other 18 cases, where at least one of the VT or ST samples differed more than a factor of 2 from CONT, there were seven where both clearly pointed in the same direction, namely houses 7–13. In these houses, retro concentrations should be preferred, taking into account the differences derived from age differences and different sampling locations within the house. From the remaining 11 houses, there are eight, namely 14–21, where VT and ST point in different directions and where one of the two is within a factor of 2 of CONT and the other within a factor of 3. These houses merit further attention. Most probably, the CONT value is not too bad as an indicator of the past radon concentration here but the retro samples urge for caution. Possibly, these houses have very different radon levels in different rooms, or changes in time have occurred. Each case should be investigated separately, checking age, origin and reliability of the retro samples in every case, which would lead us too far in this context. In the three remaining cases, namely 22–24, there is a large difference between the VT

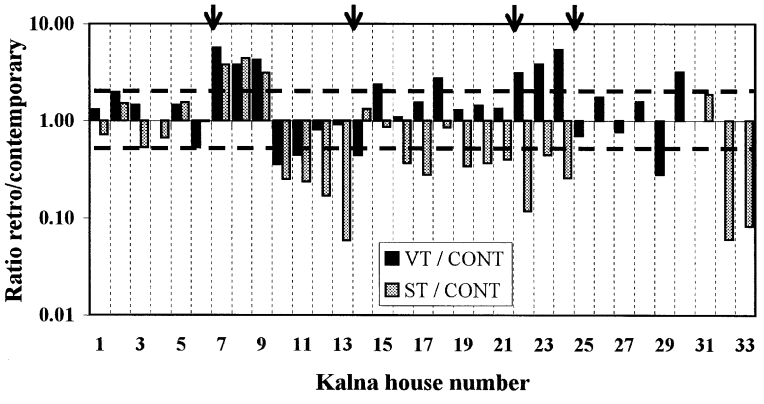

Fig. 3. Ratio of VT and ST retrospective radon estimates to contemporary radon concentrations in Kalna. The dashed lines indicate 0.5 and 2.

and ST results, more than a factor of 10. Mostly, some reasonable explanation can be found for these anomalies. Just to give some examples, for house number 22, the VT sample was seen to be very dirty and might have suffered from direct penetration of radon decay products, yielding a value that is too high. The related ST sample was stated to be 80 years old, which is very old, so some loss of radon decay products might have occurred, yielding a value that is too low. For house 23, no ready explanation is available. In the case of house 24, the VT sample was noted as doubtful since it consisted of lots of multicolored little pieces of polyester. These are sometimes found in cushions and might as well be a recycled material perhaps used earlier in other mattresses, etc. This of course renders the material worthless since age and prior radon history are unknown. The ST seemed rather acceptable in this case and seems to indicate that the past radon concentration was about a factor of 3 lower than it is today. For 25–33 unfortunately, only one of both retro results was available, but here also, only five out of nine CONT results agree within a factor of 2 with the retro result.

The Gornja Stubla case shows a special behaviour. The average CONT here was 767 Bq/m^3, varying between 300 and 1800 Bq/m^3. All retro samples were fairly new, 8.5 years on the average for VT and 19 years for ST. Fig. 4 shows that almost all VT results are around or slightly above CONT, whereas almost all ST results are a factor of 2–4 lower than CONT. The only exception is house number 10. As it happens here, both VT and ST were 20 years old. In all other cases, the VT samples were a lot younger than ST. From those 10 houses, it would seem that during the last say 5 years or so, radon concentrations have increased by about a factor of 5. This should be confirmed by further comparisons between VT and ST in other houses in the region. Such behaviour has been observed in other cases, mostly as a result of ceasing mine activities. However, in a rural community, it is a strange and so far unexplained phenomenon.

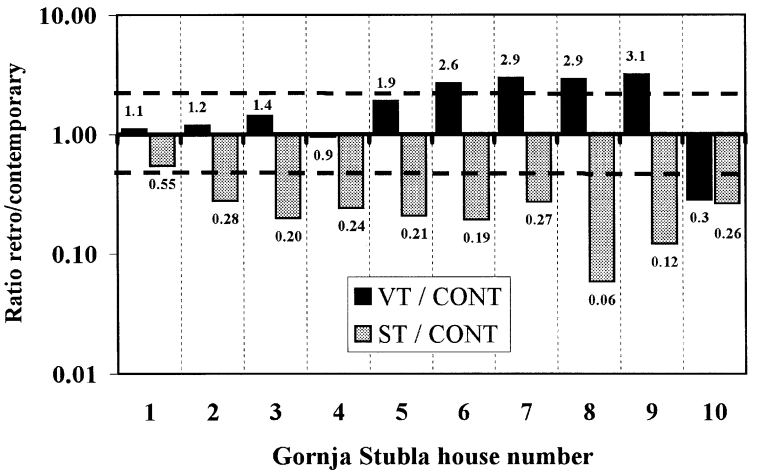

Fig. 4. Ratio of VT and ST retrospective radon estimates to contemporary radon concentrations in Gornja Stubla. The dashed lines indicate 0.5 and 2.

4. Conclusion

From this field study, it can be seen that even in stable rural communities, great caution should be taken when using 1-year contemporary radon measurements as representative for a lifetime. It is not valid, not even in 50% of the considered cases, as was seen even from the results of a retro sampling campaign that evolved as it went along. These results clearly show that a carefully planned retro sampling campaign relying on the experience gained during this one would be a considerable asset in any future or past epidemiological study. The combination of VT and ST sampling has proven to be very useful, and the fact that it makes field work more difficult does not outweigh the even simpler fact that if you want to asses radon-related health risks, you will need reliable long-term radon data.

Acknowledgements

The authors wish to acknowledge the people of Kalna, Gornja Stubla and Uzice who kindly opened their houses for this field campaign. We also acknowledge the laboratories at SSI (Sweden) and NRPA (Norway) for some of the ST results from the SINI intercomparison that were used in this article.

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High natural radiation exposure in radon spa areas: a detailed field investigation in Niška Banja (Balkan region)☆

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Abstract

The measurement campaigns have been done in the rural community of Niška Banja, a spa town located in southern Serbia, to evaluate population exposure to natural radioactivity. After a screening survey in 200 houses, annual radon and thoron concentrations were measured in 34 houses, and in 2004 a detailed investigation was carried out at six houses with elevated indoor radon concentrations. The paper presents the results of these detailed measurements. The complementary techniques were applied to determine radon and thoron concentrations in indoor air, in soil gas, radon exhalation from soil, soil permeability, and indoor and outdoor gamma doses. Soil and water samples were collected and analysed in the laboratory. Indoor radon and thoron concentrations were found to be more than 1 kBq m^{-3} and 200 Bq m^{-3}, respectively. Extremely high concentrations of soil-gas radon (>2000 kBq m^{-3}) and radon exhalation rates

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(1.5 mBq $m^{-2} s^{-1}$) were observed. These results will be utilised to set up the methodology for a more systematic investigation.

Keywords: Radon spa; Radon; Thoron; Soil; Exhalation; Gamma dose rate

1. Introduction

In the period of 1997–1999, an indoor radon survey was carried out in some regions of Yugoslavia where high radon concentration was suspected (Paridaens et al., 2000, 2001, 2002; Žunić et al., 1999, 2001, 2005). The contemporary and retrospective, both surface and volume trap techniques were applied. Attempts were made in investigated areas to identify rural populations receiving elevated natural exposures that might serve as potential cohorts for a planned future health study. Radon concentration was measured in 210 houses by using CR-39 detectors, with a total of 1443 measurements. Detectors were deployed in four seasons in living room and bedroom of each house, wherever it was feasible. Moreover, retrospective methods were used to measure the activity of radon, long-lived ^{210}Po progeny, on glass and on porous spongy household material. Two high radon-level areas were found at Gornja Stubla (Kosovo) and Kalna (east Serbia), with about 3000 and 500 inhabitants, respectively. The annual effective doses in these areas, estimated according to the ICRP-65 methodology (ICRP, 1993), amounted to 16.2 mSv and 6.8 mSv, respectively.

In a further search (2000–2003) for high radon levels, attention was directed toward spas based on thermal and mineral water, as places where a wide area may be subjected to elevated radon levels because previous surveys in similar areas of other countries have shown high radon concentration (Cosma et al., 1996; Pohl-Rüling and Hofmann, 2002). In Serbia and Montenegro there are a number of thermal and mineral springs, of which around 40 are used as spas for balneological purposes, since the Roman Empire Age.

In 2000, the first indoor radon measurements were conducted in Niška Banja (a spa town located in southern Serbia) using charcoal canisters by the Institute of Health Protection of Workers, Niš, Serbia (Manic et al., 2006). Although the measuring sites were not selected on the basis of a defined sampling scheme that took into account geology, hydrology or building material, it appeared clearly that parts of the town were highly exposed to radon. In several rooms, indoor radon concentration was found higher than 10 kBq m^{-3}.

In order to give a more comprehensive evaluation of exposure and to identify sources of very high radon levels in Niška Banja, the detailed research was undertaken in June 2004. Six dwellings with the highest indoor radon concentration were chosen for the measurements. The complementary techniques were applied to determine instantaneous and average radon and thoron concentrations, radon concentration in soil gas, radon exhalation from soil, soil permeability and gamma dose rate. In addition, soil and water samples were collected for further chemical and spectroscopic analyses. The paper describes the results of this detailed investigation in chosen high radon houses in Niška Banja town.

2. Geology of the area

In the area of the Carpathian–Balkan arc (south-eastern part of Serbia) many thermal and mineral springs occur. Their existence is enabled due to the complicated geological-tectonic

factors. Total mineralization of these springs is less than 1 g dm^{-3}. Discharge varies during the year from 0.04 $m^3 s^{-1}$ to 5 $m^3 s^{-1}$, temperature ranges from 12 °C to 39 °C. As temperature slides down, the radioactivity of water decreases and it ranges from 3.5 Bq dm^{-3} to 130.8 Bq dm^{-3} (Aksin et al., 1976).

From the geological point of view, Niška Banja town is located in the Quaternary alluvium of River Nišava along the contact of Koritnik limestones and thick strata of travertine (spring sediments) – also known as ''bigar'' in Serbian literature. This contact is also known as ''spa fault'' which enabled the formation of a number of springs out of which the most important ones are Glavno Vrelo and Suva Banja (Vujisić et al., 1980). According to more recent data (Milivojevic, 1991) ''spa fault'' (term very often used in earlier period) is brought up in question. Milivojevic (1991) states that Niška Banja represents a specific type of limestone convective systems built up of fractured Jurassic and Cretaceous limestones, open on two sides for feeding up with atmospheric water. These springs are thermal-radioactive springs and according to hydrogeological classification they are designated as ''thermo-karstic springs''.

From the point of view of geotectonic characteristics, Niška Banja town belongs to the Gornjacko-Suva Planina zone. The whole zone represents the big anticline of Suva Planina mountain, the most distinctive fold in whole eastern Serbia. General direction of anticline axes is NW-SE. It is cut with local transversal faults, and with movement along these. A lot of secondary faults in direct vicinity of Niška Banja, normal to the anticline axes of Suva Planina mountain, have a direction of SW-NE.

3. Measurement techniques

3.1. Long-term radon and thoron concentrations in indoor air

Long-term radon (and thoron) concentration in indoor air was measured by using two different alpha track detectors (polycarbonate and CR-39) enclosed in diffusion chambers.

UFO passive detector can measure both radon and thoron concentrations (Doi et al., 1992), and consists of two hemispheres with one circle shaped polycarbonate film with 50 mm diameter (IUPILON) in each hemisphere. In the upper, larger hemisphere (110 mm diameter), both radon and thoron can enter through filters, so the film detects radon, thoron and their progenies produced inside, whereas only radon succeeds to enter the bottom, smaller hemisphere (70 mm diameter), so the film detects only radon and its daughters produced inside. Thoron concentration is obtained by difference, using appropriate calibration factors. After exposure, detector films were etched and then scanned for track density measurement by using a microfiche reader (Canon, Microprinter 60). Upper exposure limit of UFO detector is 680 kBq m^{-3} h.

Radon concentration was measured also with a commercial system (Radosys) which uses (1×1 cm) CR-39 as detector material, enclosed in a small cylindrical (5 cm height, 3 cm diameter) diffusion chamber. Detectors were etched in 6.2 M NaOH at 90 °C for 4.5 h, washed in fresh water for 10 min and dried overnight. The tracks produced by the alpha particles were counted with an automatic set-up composed of an optical microscope connected to a CCD camera controlled by a personal computer. Saturation occurs at exposures higher than about 10 MBq m^{-3} h.

After selecting the locations for the measurements, UFO and Radosys detectors were deployed at a distance of 20–30 cm from the wall and 1–2 m above the floor.

3.2. Short-term radon and thoron concentrations in indoor air

In the second step of the study (June 2004), short-term radon concentration was measured by active methods, using scintillation cells, ionization chamber (AlphaGuard PQ 2000, produced by Genitron, Germany), and instruments with semiconductor detectors (EQF 3020, EQF 3020-2 and RTM 2010-2, SARAD, Germany). The owners of the houses were asked to keep the selected rooms closed overnight prior to the measurements. Air samples were collected directly into the alpha scintillation cells (Vaupotič et al., 1992) and the activity was measured with a PRM-145 alpha counter (AMES, Slovenia) after 3 h, when radon and its short-lived decay products reached radioactive equilibrium. In some rooms, the other active instruments were used for continuous monitoring of the radon concentration with a frequency from once in an hour to once in 2 h.

In order to comply with QA/QC requirements, alpha scintillation cells and portable measuring devices have been regularly checked at the inter-comparison experiments organised annually by the Nuclear Safety Administration at the Slovene Ministry of the Environment and Spatial Planning (Križman, 2001).

3.3. Radon and thoron concentrations in soil

The measurement set-up consisted of the AlphaGUARD PQ 2000 PRO (AG) radon monitor, a soil-gas probe and a pump (AlphaPUMP). A 1-m deep hole was drilled and the soil gas was pumped into the ionization chamber of the AlphaGUARD (Fig. 1). The total concentration of both isotopes (^{222}Rn and ^{220}Rn) was measured in this way. In order to determine radon (^{222}Rn) concentration only, the second measurement was carried out after filling the ionization chamber with soil gas and keeping it closed tightly for about 10 min – time needed for thoron (^{220}Rn) to

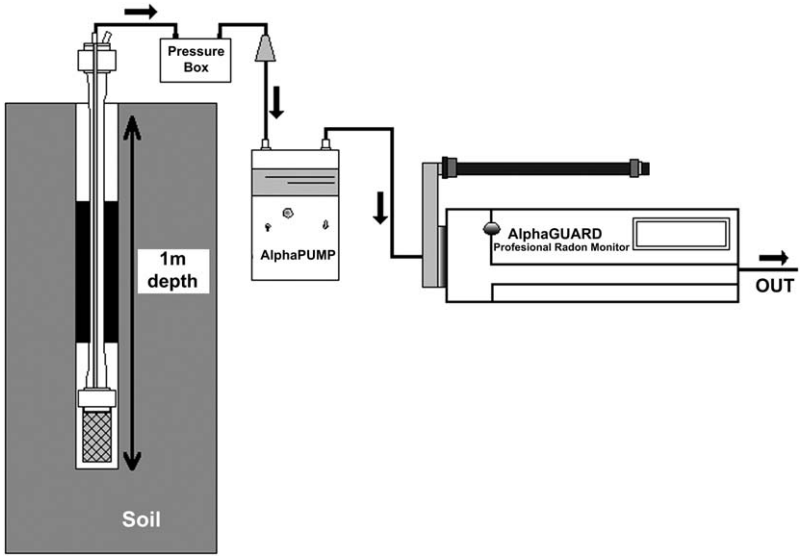

Fig. 1. Schematic set-up for radon and thoron in soil-gas measurements.

decay. The concentration of thoron was determined as a difference between the first and the second measurement.

3.4. Radon exhalation rate from soil

Radon exhalation rate was measured ''in situ'' using the Exhalation Box with special valves (Swagelock type), a pump and the AG radon monitor which registers the increasing radon concentration inside the Exhalation Box (Fig. 2). The air was circulated in the closed circuit for about 90 min. The value of exhalation rate has been determined from the slope of linear fit to the experimental data.

3.5. Radon (^{222}Rn) and radium (^{226}Ra) concentrations in water

The AG radon monitor with the additional equipment AquaKIT (a set of special glass vessels and pipes) was used for the measurements of radon concentration in water. Fig. 3 shows the set-up for this type of radon measurements. More details of the method can be found elsewhere (Kochowska et al., 2004).

For ^{226}Ra analysis in water, the sorption–emanation method was used. A water sample (3–5 dm^3) is collected in a polyethylene bottle and acidified with nitric acid to a pH below 2. In the laboratory, the sample is passed through a column filled with 50 cm^3 of cationic exchange resin to adsorb radium. The resin is purged with aged nitrogen gas and closed for 10–20 days for radon (^{222}Rn) growth from the radium adsorbed. Then, the column is connected to a vacuum system and the resin is purged with aged nitrogen gas to expel radon from the sample and transfer it into a scintillation cell: carrier gas with radon is first dried in a trap with concentrated sulphuric acid and then passed through a trap filled with glass beads and cooled with liquid nitrogen, in which radon is separated. On warming up the beads, radon evaporates and is filled into an evacuated alpha scintillation cell. After 3 h, when a radioactive equilibrium between radon and its short-lived decay products is reached, the cell is counted in an alpha scintillation counter and ^{226}Ra activity concentration calculated from the ^{222}Rn activity measured.

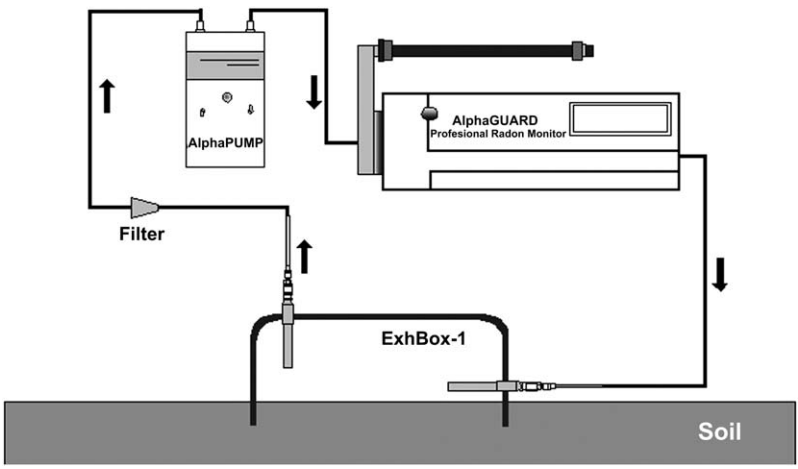

Fig. 2. Schematic set-up for radon exhalation rate measurements.

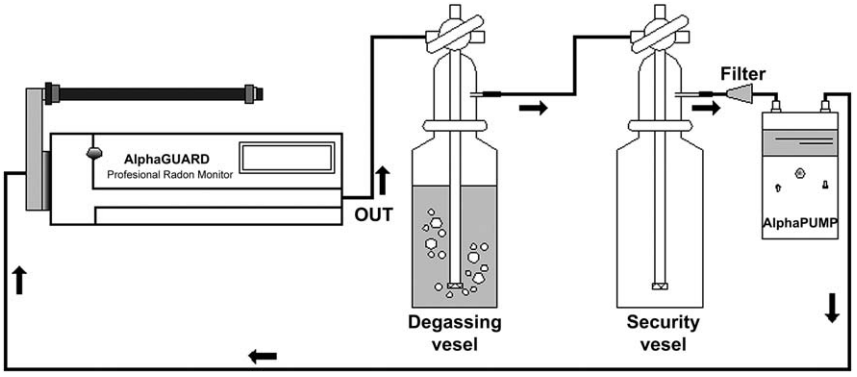

Fig. 3. Schematic set-up for measurements of radon in water.

3.6. Indoor and outdoor gamma dose rates

Gamma dose rate was measured using two devices: the AlphaGUARD radon monitor with a GM tube and a portable (Eberline, USA) scintillation counter ASP-1. The registration of outdoor gamma dose rate was performed at 0–20 cm above the ground level. Indoor gamma dose rate was measured in the same rooms where radon was measured. Typical time of the measurements was 2–3 h.

3.7. Soil permeability

The measurement set-up consisted of the Pressure Gauge VDPT-10S (range 0–10 kPa), a rotameter (0–1.0 $dm^3\ min^{-1}$), a soil-gas probe and a pump (AlphaPUMP). A 1-m deep hole was drilled and the air was pumped into the hole through the soil-gas probe (Fig. 4). In this way, the pressure difference and a flow rate was measured. The modified equation from Fick's law of diffusion was used to calculate the permeability:

$$k = \mu \frac{1}{W} \frac{Q}{\mathrm{dP}}$$

where k is permeability (m^2), μ is dynamic viscosity (Pa s), W is shape coefficient for soil probe (m), Q is flow rate ($m^3\ s^{-1}$), and dP is pressure difference (Pa). The shape coefficient, W, is a semi-empirical parameter determined in the calibration device. It depends on the flow rate and is different for different soil-gas probe designs.

3.8. Concentration of natural radioactive elements in soil

The soil samples were collected from the study area for the determination of concentrations of natural radioactive elements – ^{226}Ra, ^{228}Th and ^{40}K. They were taken from the depth of 0.6–1.0 m while drilling holes for the radon measurements in soil. In the laboratory the soil samples were crushed and dried (105 °C) before putting into Marinelli beakers. In order to reach the radioactive equilibrium between ^{226}Ra and ^{222}Rn the samples were kept closed for about 4 weeks. Then they were measured by means of a low-background spectrometry with $3'' \times 3''$ NaI(Tl) scintillation detector. Radium ^{226}Ra was determined using 1.76 MeV peak

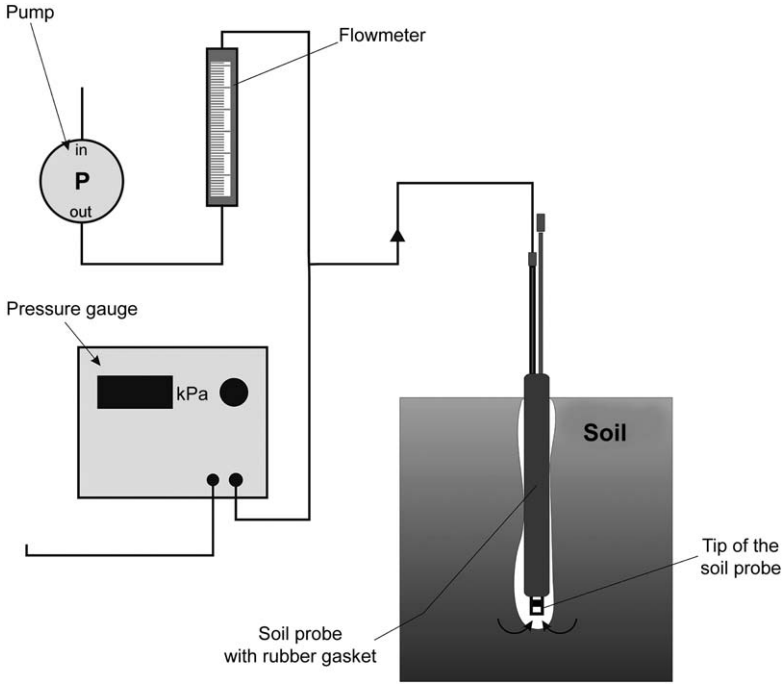

Fig. 4. Schematic set-up for soil permeability measurement.

(from ^{214}Bi decay) and thorium ^{228}Th using 2.62 MeV peak (from ^{208}Tl decay). Time of the measurement was from 10,000 to 30,000 s for each sample. The concentrations of natural radioisotopes were calculated using a ''three-window'' method. The IAEA materials (RGU-1, RGTh-1, RGK-1) were used as standards.

4. Results

The results of the indoor and outdoor natural radiation measurements at six houses in Niška Banja town are presented in Tables 1–4. The uncertainties given in the tables are total measurement errors due to both statistical and experimental reasons.

Table 1
Results of outdoor radon measurements

Code	Concentration in soil		Radon exhalation rate (mBq m^{-2} s^{-1})	Soil permeability (m^2)
	Radon (kBq m^{-3})	Thoron (kBq m^{-3})		
H1	63.7 ± 2.2	20 ± 4	30 ± 6	$1.11\times10^{-12} \pm 3.41\times10^{-14}$
H2	77.5 ± 2.6	5 ± 4	96 ± 18	$2.77\times10^{-12} \pm 1.70\times10^{-13}$
H3	>2000[a]	Not measurable	450 ± 71	$2.13\times10^{-12} \pm 1.01\times10^{-13}$
H4	1300 ± 45	0 ± 62	671 ± 86	$4.51\times10^{-12} \pm 2.89\times10^{-12}$
H5	980 ± 33	20 ± 50	1542 ± 354	$2.67\times10^{-12} \pm 4.38\times10^{-13}$
H6	74.3 ± 2.4	46 ± 5	57.9 ± 11.9	$1.41\times10^{-12} \pm 2.01\times10^{-13}$

[a] Out of AlphaGUARD PQ 2000 PRO upper detection limit.

Table 2
Results of indoor radon (Rn) and thoron (Tn) concentrations obtained by passive and active methods

Code	Method	Type of detector	Start date	End date	Rn (Bq m^{-3})	Tn (Bq m^{-3})
H1	Track etched	RADOSYS	27.11.03	26.03.04	215 ± 20	–
		UFO	27.11.03	26.03.04	210 ± 28	20 ± 26
			26.03.04	12.06.04	221 ± 30	18 ± 28
			12.06.04	21.09.04	103 ± 9	28 ± 32
			21.09.04	17.11.04	180 ± 15	0 ± 55
	Instantaneous	SC	12.06.04	12.06.04	450 ± 20	–
	measurements	AG	–	–	–	–
		SARAD	–	–	–	–
H2	Track etched	RADOSYS	27.11.03	26.03.04	262 ± 26	–
		UFO	27.11.03	26.03.04	250 ± 33	26 ± 31
			26.03.04	12.06.04	230 ± 31	2 ± 29
			12.06.04	21.09.04	92 ± 8	9 ± 29
			21.09.04	17.11.04	120 ± 11	0 ± 39
	Instantaneous	SC	15.06.04	15.06.04	500 ± 25	–←
	measurements	AG	14.06.04	15.06.04	1150 ± 70	–←
		SARAD	14.06.04	15.06.04	1440 ± 130	–←
H3	Track etched	RADOSYS	11.03.03	29.05.03	4010 ± 390	
			29.05.03	17.10.03	1020 ± 100	–
			17.10.03	19.01.04	>4400	
			19.01.04	25.03.04	>6300	
		UFO	27.03.03	29.05.03	>1460	–
			29.05.03	17.07.03	>624	–
			17.07.03	17.10.03	>620	–
			17.10.03	19.01.04	>2130	–
	Instantaneous	SC	14.06.04	14.06.04	5100 ± 75	–←
	measurements	AG	–	–	–	–←
		SARAD	13.06.04	14.06.04	3050 ± 280	–←
H4	Track etched	RADOSYS	11.03.03	29.05.03	2400 ± 220	
			29.05.03	17.10.03	1410 ± 140	–
			17.10.03	19.01.04	>4400	
			19.01.04	25.03.04	>6300	
		UFO	27.03.03	29.05.03	>1240	–
			29.05.03	17.07.03	760 ± 55	320 ± 210
			17.07.03	17.10.03	>820	–
			17.10.03	19.01.04	>960	–
	Instantaneous	SC	12.06.04	12.06.04	775 ± 30[a]	–←
	measurements	AG	13.06.04	14.06.04	5700 ± 200	–←
		SARAD	–	–	–	–
H5	Track etched	RADOSYS	11.03.03	29.05.03	730 ± 66	
			29.05.03	17.10.03	1560 ± 160	–
			17.10.03	19.01.04	>4400	
			19.01.04	25.03.04	>6300	
		UFO	27.03.03	29.05.03	>1240	–
			29.05.03	17.07.03	>829	–
			17.07.03	17.10.03	>1150	–
			17.10.03	19.01.04	>1310	–
	Instantaneous	SC	15.06.04	15.06.04	5780 ± 75	–←
	measurements	AG	–	–	–	–←
		SARAD	15.06.04	16.06.04	1570 ± 140	–←

Table 2 (*continued*)

Code	Method	Type of detector	Start date	End date	Rn (Bq m^{-3})	Tn (Bq m^{-3})
H6	Track etched	RADOSYS	27.03.03	29.05.03	212 ± 17	–
			29.05.03	17.10.03	100 ± 10	–
			17.10.03	25.03.04	617 ± 60	–
		UFO	27.03.03	29.05.03	235 ± 19	79 ± 70
			29.05.03	17.07.03	90 ± 10	51 ± 34
			17.07.03	17.10.03	158 ± 13	1 ± 46
			17.10.03	19.01.04	464 ± 34	127 ± 130

SC, alpha scintillation cells ($V = 0.7$ dm^3); AG, professional radon monitor AlphaGUARD (Genitron); SARAD, radon and radon decay products monitors; –, not measured.

The meteorological parameters (air temperature, pressure and relative humidity) were registered by the AG radon monitor during the outdoor radon measurements (radon in soil gas and radon exhalation rate) because it is known that they can influence radon concentration. Therefore it seems important to know under what conditions the short-term radon measurements were carried out. The average values of air temperature, air pressure and relative air humidity ranged from 24.0 °C to 32.9 °C, 986.4 hPa to 996.1 hPa and 50% to 75%, respectively.

The results of soil-gas radon and thoron concentrations, radon exhalation rate and the soil permeability are shown in Table 1. At one measurement place (H3) the soil-gas radon concentration exceeded the upper detection limit of the detector (2 MBq m^{-3}). At other two locations (H4 and H5) soil-gas radon concentration was also recorded which was very high, reaching almost 1 MBq m^{-3}. The radon exhalation rate from soil again showed very high values at locations H3, H4 and H5. Soil permeability at all locations was found nearly the same ranging from 1.11×10^{-12} to 4.51×10^{-12} m^2.

The results of indoor radon measurements are shown in Table 2. The measurements using passive radon detectors (RADOSYS and UFO types), whose exposure time varied from 2 to 5 months, were made during the years 2003 and 2004. In June 2004, the ''instantaneous'' (actually short-time) measurements of indoor radon concentration were also performed in the same houses (except house H6) using scintillation cells and active radon monitors (AlphaGUARD and SARAD). The highest radon concentration was found (both by passive and active methods) in the houses H3, H4 and H5. The results obtained using passive techniques ranged from 620 Bq m^{-3} to 4010 Bq m^{-3}. The values of radon concentration could have been even higher because foils of passive detectors were sometimes saturated. The results of short-term measurements obtained using active devices varied from 775 Bq m^{-3} to 5780 Bq m^{-3}. These results correlate to the above-mentioned results of radon in soil measurements. Soil gas and indoor radon concentrations were found to be higher at the same locations, indicating that soil was the main source of radon.

Table 3
Results of gamma spectroscopic measurements of soil samples from Niška Banja

Sample code	^{40}K (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{228}Th (Bq kg^{-1})
H1	430 ± 20	45 ± 2	41 ± 2
H2	330 ± 20	260 ± 10	24 ± 1
H3	90 ± 6	820 ± 40	11 ± 1
H4	440 ± 30	610 ± 30	43 ± 2
H5	310 ± 20	1150 ± 60	33 ± 2
H6	410 ± 20	120 ± 10	36 ± 2

Table 4
Results of radium and radon in water measurements

Sample code	^{226}Ra concentration (Bq dm^{-3})	^{222}Rn concentration (Bq dm^{-3})
W1	0.650 ± 0.015	23 ± 2
W2	0.215 ± 0.010	460 ± 20
W3	0.550 ± 0.020	0.2 ± 0.4
W4	Not measured	570 ± 20
W5	Not measured	510 ± 20
W6	0.046 ± 0.003	0.5 ± 1.6
W7	0.033 ± 0.002	3.0 ± 0.5
W8	0.460 ± 0.015	Not measured
W9	0.038 ± 0.003	Not measured
W10	0.360 ± 0.010	Not measured
W11	0.315 ± 0.010	Not measured
W12	0.039 ± 0.003	Not measured

The values of indoor gamma dose rate (using ASP-1) ranged from 110 nSv h^{-1} (H2) up to 550 nSv h^{-1} (H5). The outdoor measurements (using AG and ASP-1) showed the results ranging from 128 nSv h^{-1} (H6) to 276 nSv h^{-1} (H3). The results of both devices were in good agreement – the maximum discrepancy amounted to 20%.

The results of gamma spectroscopic measurements of soil samples are presented in Table 3. The highest radium (^{226}Ra) content (over 800 Bq kg^{-1}) was found at locations H3, H4 and H5, where the concentration of radon in soil gas also reached the highest values. The potassium (^{40}K) and thorium (^{228}Th) contents in the soil varied from 309 Bq kg^{-1} to 436 Bq kg^{-1} and from 24 Bq kg^{-1} to 43 Bq kg^{-1}, respectively. In location H3, much lower concentrations of ^{40}K (86 Bq kg^{-1}) and ^{228}Th (Bq kg^{-1}) were observed.

The values of radium (^{226}Ra) and radon (^{222}Rn) contents in 12 water samples (W1–W12) are shown in Table 4. The samples W2–W5 and W8–W12 were taken from wells located in Niška Banja. W6 and W7 are groundwater samples from supplies of drinking water for Niš Valley, located 40 km east of Niška Banja. W1 sample was taken from one of the thermal springs in Niška Banja. Half of the total number of measured samples showed the values of radium concentration higher than the proposed EPA drinking water standard for total radium (0.185 Bq dm^{-3}). It can be also seen that the contents of radium and radon in some samples are not in agreement (W1, W3). It is mainly because radon content in water depends on the radium concentration in rock material of the aquifer rather than on radium concentration in water. Another reason for this disagreement is improper collection of water samples. It is well known that the manner of collecting water sample is essential for proper determination of radon concentration. Sometimes this is not possible – in Niška Banja water samples were taken from taps or "open" streams of thermal water. Therefore radon could have escaped easily before closing the containers and in this case the results are underestimated.

5. Discussion and conclusions

On the basis of field measurements the following considerations can be drawn.

1. Radon concentrations were found to be very high in soil gas, water and indoor air in Niška Banja town and for the soil gas concentrations have exceeded the upper detection limit (>2 MBq m^{-3}) of the detector at some places. The concentration of thoron in soil gas was also recorded high (20–46 kBq m^{-3}).

2. The observed high radon exhalation rate from the soil may result in the high radon concentration in the houses of Niška Banja. However, the soil permeability was found similar in all locations.
3. Many long-term measurements of indoor radon concentration with passive detectors (CR-39 and polycarbonates) are reported in Table 2 as lower limit due to detector saturation. Future measurements in similar situations need to be done with track detectors in low efficiency configurations in order to avoid saturation. Uncertainties of thoron concentration from UFO detectors are too high. UFO detector is discriminative type of passive detector. Therefore, radon concentration uncertainty is included in thoron concentration uncertainty. When the radon concentration is much higher than the thoron concentration, radon concentration uncertainty and therefore thoron concentration uncertainty are higher than thoron concentration itself.
4. The measured high radon contents in soil, water and air are due to the presence of high radium contents in the soil and water. However, thorium and potassium contents were found within the average level. The high radium contents in the soil and the groundwater may be due to the presence of uranium mineralization in the area. It is also possible that radium has been transported to Niška Banja area from its parent nuclei after the leaching. The presence of local faults in the study area may also enhance the radon emanation from the soil.
5. The levels of indoor radon recorded by different passive techniques (RADOSYS and UFO) were generally found in good agreement when comparing the similar periods of exposure time.

In conclusion, these results show that radon concentration in indoor air and soil of Niška Banja can reach extremely high values, correlated with high radium content in soil and high radon exhalation from soil. More complex research is required to explain the existence of higher thoron levels in soil in places where thorium content amounts to the average value. These results will be utilised to set up the methodology for a more systematic investigation to assess the risk due to high radiation exposure to the general population in Niška Banja town. Guidelines on the use of spa water will also be needed in order to avoid high exposures.

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Radon survey in the high natural radiation region of Niška Banja, Serbia

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Abstract

A radon survey has been carried out around the town of Niška Banja (Serbia) in a region partly located over travertine formations, showing an enhanced level of natural radioactivity. Outdoor and indoor radon concentrations were measured seasonally over the whole year, using CR-39 diffusion type radon detectors. Outdoor measurements were performed at 56 points distributed over both travertine and alluvium sediment formations. Indoor radon concentrations were measured in 102 living rooms and bedrooms of 65 family houses. In about 50% of all measurement sites, radon concentration was measured over each season separately, making it possible to estimate seasonal variations, which were then used to correct values measured over different periods, and to estimate annual values. The average annual indoor radon concentration was estimated at over 1500 Bq/m^3 and at about 650 Bq/m^3 in parts of Niška Banja located over travertine and alluvium sediment formations, respectively, with maximum values exceeding 6000 Bq/m^3. The average value of outdoor annual radon concentration was 57 Bq/m^3, with a maximum value of 168 Bq/m^3. The high values of indoor and outdoor radon concentrations found at Niška Banja make this region a high

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natural background radiation area. Statistical analysis of our data confirms that the level of indoor radon concentration depends primarily on the underlying soil and building characteristics.

Keywords: Radon; Survey; Natural radiation; Travertine

1. Introduction

Niška Banja is located in the quaternary alluvium of the River Nišava along the contact of the Koritnik limestone and a thick strata of groundwater spring deposits (travertine) of South East Serbia. The formation of travertine in the presence of high radium concentrations in the spring water of the Niška Banja region is similar to that of the well-known high natural background radiation areas of Ramsar in Iran. According to Ghiassi-Nejad et al. (2002), the high natural background radiation in Ramsar results primarily from very high concentrations of ^{226}Ra in hot spring waters. The average ^{226}Ra concentration measured in soils around Niška Banja is about 900 Bq/kg (Mazur et al., 2005). Radium-enriched travertine is a source of radon and, due to its porous structure, permits radon migration, resulting in a high level of radon exposure in the dwellings of this region.

Early in 2000, screening indoor radon measurements using charcoal canisters were conducted in Niška Banja by the Institute of Occupational Health, Niš, Serbia (Manic et al., 2006). Following a request from the local community to confirm those results, in March 2003 a second survey was undertaken, using long-term measurement techniques. This survey was carried out in the framework of an international project coordinated by the Institute of Nuclear Sciences Vinča, Belgrade (Serbia). The project includes a detailed field investigation of Niška Banja spa area (Žunić et al., 2006). The present paper is devoted to report and discuss the results of long-term measurements of outdoor and indoor radon concentrations and of the analysis of factors affecting these values.

2. Materials and methods

2.1. Geology

The Niška Banja region lies in the mountain range known as the Karpato-Balkan arch. Niška Banja is located in the eastern part of the Niš neogene basin, on the southern slopes of the Koritnik mountain. A formation of particular interest in our case is quaternary travertine, a sediment formed as a result of carbonate precipitation from spring waters. Generally, travertine is formed when water, saturated with carbonate, flows from the spring, its pressure and temperature drops, and the solubility of carbonate decreases. At this stage, carbonate precipitates and travertine is formed. A travertine plateau occupies the central parts of Niška Banja.

During the period from 1986 to 1987 a drill bore hole was made in the southern part of the town of Niška Banja. From the core, the composite summary geological profile under the town was established as follows: 0–135 m clay and marl sediments – Neogene; 135–220 m flysch sediments – Lower Cretaceous; 220–350 m clay rich Lower Cretaceous limestone; 350–470 m compact Lower Cretaceous limestone; 470–510 m limestone saturated with mineral water (Milivojević, 1991).

2.2. Measurement sites and techniques

Outdoor radon measurements were performed at 56 points evenly distributed over the territory of Niška Banja (in the gardens, 1 m above soil, 2–7 m away from the house walls). For indoor radon measurements 16 houses were randomly selected from the 200 previously surveyed measurement points (Manic et al., 2006). Additionally, 49 houses not previously surveyed, in the area expected to be radon-prone, were selected, giving a total of 65 houses surveyed. All selected houses are single-family, one- or two-floor buildings. In each house, radon concentration was generally measured in the living room and bedroom, giving a total of 102 rooms surveyed. The outdoor and indoor measurements were conducted over the period from March 2003 to January 2006 during different seasons in order to account for seasonal variations of radon concentration. Measurement periods were denoted as winter, spring, summer or autumn according to the mid-date of the measurement period.

Radon concentration measurements were performed using 1 cm^2 CR-39 detectors, enclosed in small cylindrical (5 cm height, 3 cm diameter) diffusion chambers. Detectors were etched in 6.2 M NaOH at 90 °C for 4.5 h, washed in clean water for 10 min and dried overnight. Tracks on the CR-39 were counted with an automatic setup consisting of an optical microscope connected to a CCD camera controlled by a personal computer. Durations of exposure (typically 3–5 months) were chosen to reduce the probability of saturating the detector response. Nevertheless, in some cases, especially for indoor winter measurements, detector saturation did occur at exposures exceeding 10 MBq/m^3 h.

A questionnaire was developed and filled in by trained technicians for every house surveyed, with information provided mainly by its inhabitants. Apart from house identification data (owner's name, address, GPS coordinates etc.), the following data were obtained: floor level, wall material type, wall surface treatment, year of building construction, facade type, number of smokers and number of cigarettes they smoked per day, source of tap water, floor type, presence of hydro-isolation, and types of chimney, ventilation, heating, and of underlying rock.

Additionally, outdoor and indoor gamma dose rates were measured, in terms of kerma in air, with high-sensitive LiF:Mg,Cu,P thermoluminescence (TL) detectors (Budzanowski et al., 2004). The TL dosimeters were exposed at 55 outdoor and 55 indoor locations during 4 months over the period November 2003–April 2005. The TL-measured values of outdoor terrestrial air kerma-rates were corrected by subtracting the cosmic-ray background of 32.6 nGy/h, as calculated with the CARI-LF code for solar potential over the first months of 2004 and 2005 and the coordinates and altitude of Niška Banja (Budzanowski et al., 2004), whereas for indoor values the UNSCEAR shielding factor of 20% was used and 26.1 nGy/h was subtracted.

2.3. Statistical analysis

To investigate the different factors affecting radon levels, standard analysis of variance (ANOVA) was applied. Taking into account the lognormal distribution of indoor radon concentrations, natural logarithms of the respective values were considered in this statistical analysis. Both tests of homogeneity of means and variances (Levene's test) were performed.

3. Results and discussion

3.1. Outdoor radon concentrations

The distribution of measured values of the outdoor radon concentration was found to be close to lognormal, in each of the set of measurements performed over the given seasons. Lognormal parameters (geometric mean, GM, and geometric standard deviation, GSD) and other distribution parameters of the 122 measurements analyzed, are presented in Table 1.

Table 1
Parameters of the distributions of measured values of outdoor radon concentration

Season	N	AM (Bq/m^3)	GM (Bq/m^3)	GSD	Max (Bq/m^3)
Winter (December–February)	39	51	41	2.1	116
Spring (March–May)	30	50	40	2.1	168
Summer (June–August)	16	73	60	2.1	153
Autumn–Winter (September–February)	37	56	43	2.1	244
Annual (estimated)	56	57	49	1.8	168

N = number of measured points, AM = arithmetic mean, GM = geometric mean, GSD = geometric standard deviation.

It is interesting to note that the summer outdoor radon concentration values significantly exceeded the values measured over the remaining three seasons ($p = 0.03$), while the difference between values obtained over winter, spring and autumn was found not to be significant ($p > 0.05$). Seasonal differences of standard deviations were also tested and were found not to be significant ($p > 0.05$).

In order to estimate the annual mean outdoor radon concentration for each point, the pattern of seasonal variation of outdoor radon concentration was analyzed using measurement data from 14 points out of the total of 56 measurement points where at least two seasonal measurements (one of which was summer) were performed. Having stated the homogeneity of winter, spring and autumn values, they were combined and compared with the summer values. The correlation between values of concentration measured at given measurement points over the summer and over the remaining seasons (winter, spring and autumn) combined is close to linear (Fig. 1). Using the logarithms of radon concentration values, the slope factor was estimated at 1.35 ± 0.30 and the intercept at 7 ± 12, with a correlation coefficient $R = 0.96$. Using these parameters the annual outdoor radon concentration was estimated for each point, and the summary results presented in the last row of Table 1.

The dependence of annual outdoor radon concentration on the type of underground bedrock was tested and found to be statistically significant (Fig. 2). The geometric mean of annual outdoor radon concentration in the travertine part of Niška Banja significantly exceeds the value measured in the alluvium part (comparison of means $p = 0.006$, test of homogeneity of

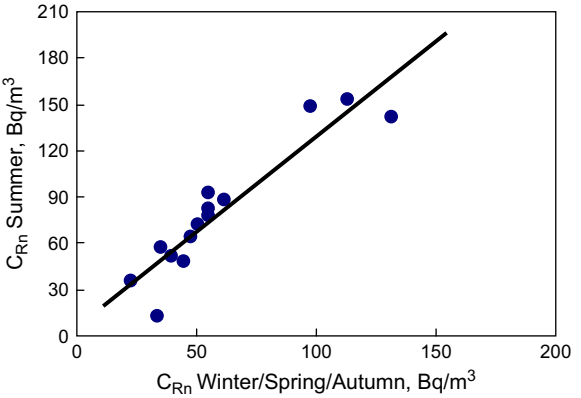

Fig. 1. Correlation between values of outdoor radon concentration measured over the summer vs values measured over the remaining seasons (winter, spring and autumn). A linear regression dependence can be fitted to this data.

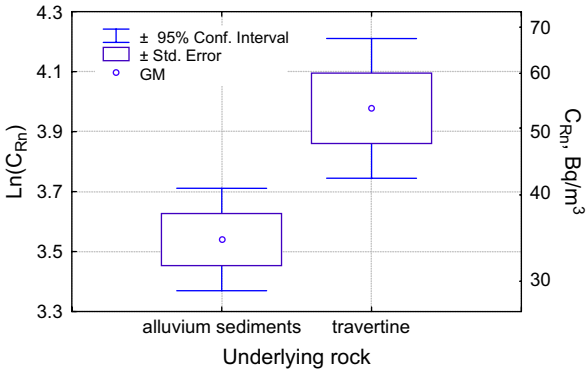

Fig. 2. Dependence of outdoor radon concentration on type of underlying bedrock (travertine: AM = 62 Bq/m^3, GM = 53 Bq/m^3, GSD = 1.8, $N = 24$ and alluvium sediments: AM = 38 Bq/m^3, GM = 34 Bq/m^3, GSD = 1.6, $N = 25$).

variances $p > 0.05$). It is worth noting that each of these average values in either part (70 and 43 Bq/m^3, respectively) considerably exceed the typical landmass worldwide value of 10 Bq/m^3 (UNSCEAR, 2000).

The correlation between the outdoor gamma dose rates and type of underlying bedrock was also tested. Similarly to outdoor radon concentrations, higher values of outdoor gamma dose rates are associated with travertine (Table 2). The average outdoor radon concentration moving from the alluvium to the travertine part increases by a factor of about 1.6 while the outdoor gamma dose rate increases by a similar factor of 1.7. Another interesting result is the significant difference between the standard deviations of outdoor gamma dose rates over travertine and alluvial terrains (test of homogeneity of variances $p = 0.044$). The higher variability of ^{226}Ra concentration over travertine indicates the higher probability of finding locally elevated radon concentrations both outdoors and indoors.

3.2. Indoor radon concentrations

3.2.1. Seasonal variation

Indoor radon concentration distribution parameters in different seasons are presented in Table 3. Within each season, indoor radon concentration follows a lognormal distribution. Out of 102 rooms, successful full year measurements were performed in 33 rooms and three

Table 2
Parameters of the distributions of measured values of outdoor and indoor gamma dose rates, by type of underlying bedrock, nGy/h

Underlying bedrock	N	AM	SD	Min	Max
Outdoor					
Alluvium sediments	28	53	12	36	104
Travertine	27	89	31	42	155
Indoor					
Alluvium sediments	26	73	26	46	155
Travertine	29	94	40	42	188

N = number of measured points, AM = arithmetic mean, SD = standard deviation.

Table 3
Parameters of the distributions of measured values of indoor radon concentration over different seasons

Season	N	AM (Bq/m^3)	GM (Bq/m^3)	GSD	Max (Bq/m^3)	%>600 (Bq/m^3)
Winter	66	1256	557	4.0	>10 000[a]	48%
Spring and Autumn	100	1185	461	4.5	>6000[a]	43%
Summer	47	485	215	3.6	2164	21%
Annual (estimated)	102	1163	529	3.9	6155	46%

N = number of measured rooms, AM = arithmetic mean, GM = geometric mean, GSD = geometric standard deviation.
[a] CR-39 detector saturation was observed at exposures above 10 MBq/m^{3}h.

or two seasonal measurements in 69 rooms. Saturation of CR-39 detectors appeared in 14 and three rooms over winter and autumn measurements, respectively. In order to estimate the annual radon concentration using incomplete data (missing or saturated measurements) a seasonal normalization approach was developed. It was found that a simple linear function best fits the correlation of pairs of indoor radon seasonal measurements. The results of regression analysis are presented in Fig. 3 using a logarithmic scale (therefore, in Fig. 3, the fitted line appears to be non-linear). The parameters of the linear function obtained for each regression are presented in Table 4. Using measured and estimated seasonal values, the annual indoor radon concentration was estimated and its distribution parameters are presented in Table 3 (last row).

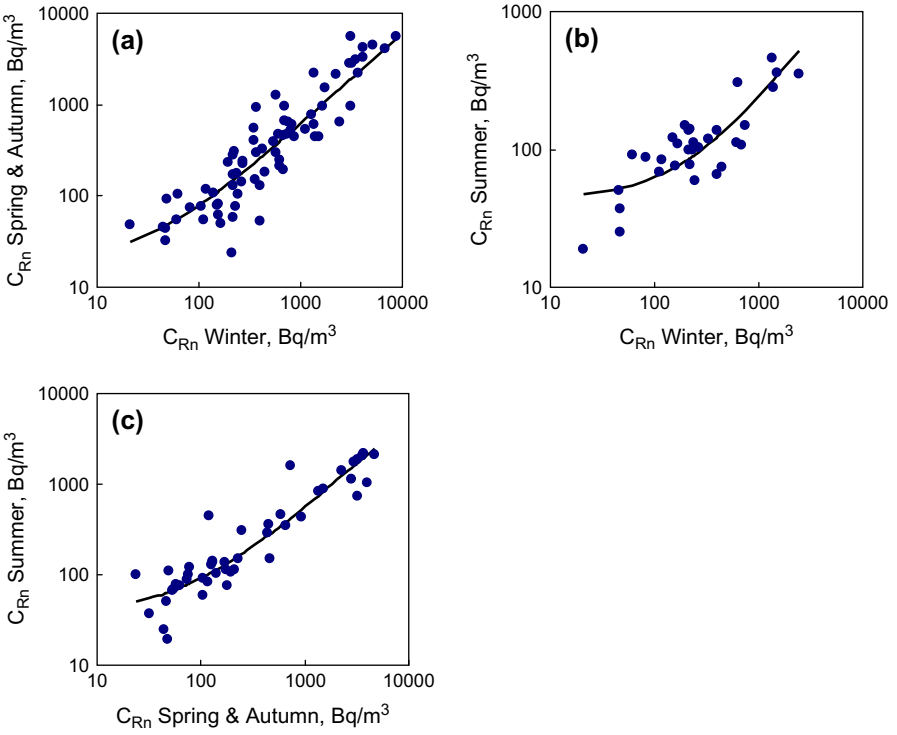

Fig. 3. Correlation between values of indoor radon concentration measured over (a) spring and autumn vs winter, (b) summer vs winter, (c) summer vs spring and autumn. The regression parameters of linear dependences fitted to these data sets are given in Table 4. Note that linear dependences are shown in doubly logarithmic plots.

Table 4
Parameters of linear regression fitted to correlated pairs of seasonal indoor radon concentrations with 95% confidence intervals (the second season is the independent parameter of the linear model)

Season pairs	Slope	Intercept (Bq/m^3)	R
Spring and Autumn vs Winter	0.60 ± 0.11	19 ± 18	0.90
Summer vs Winter	0.20 ± 0.07	43 ± 18	0.81
Summer vs Spring and Autumn	0.52 ± 0.14	39 ± 20	0.93

3.2.2. Factor analysis

In the following analysis we investigate the different factors that can affect annual indoor radon levels. Among other characteristics documented during this survey the factor ''type of underlying bedrock'' is considered as being most influential. This factor incorporates several geological and geophysical aspects of our study. Differences between geometric means of annual indoor radon concentration in dwellings situated over alluvium and travertine parts of Niška Banja are presented in Fig. 4a. Higher values of indoor radon concentration are associated with travertine ($p = 0.0003$). Although the test of homogeneity of variances shows insignificant difference ($p = 0.5$) as in the outdoor cases, a higher standard deviation is obtained for the travertine group. Higher average values and standard deviations resulted in doubling of the

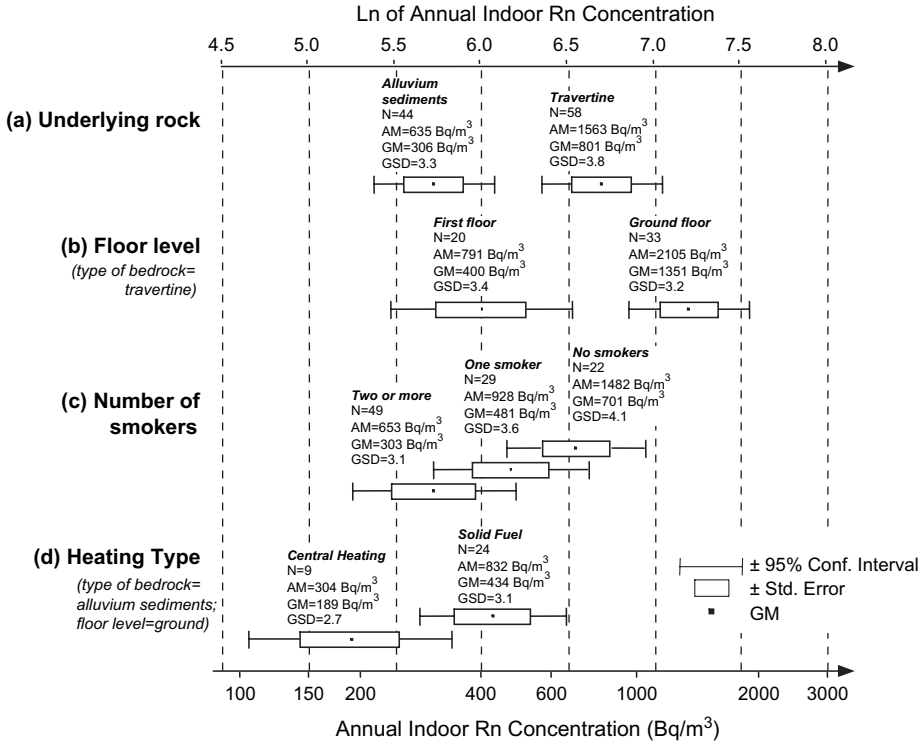

Fig. 4. Dependence of indoor radon concentration on (a) type of underlying bedrock; (b) floor (type of bedrock = travertine); (c) number of smokers; and (d) heating type (type of bedrock = alluvium sediments, floor = ground).

percentage of dwellings with indoor radon concentrations exceeding the 600 Bq/m^3 level in the travertine part (59%), compared with the alluvium part of Niška Banja (29%). The indoor gamma dose rate correlates with the type of underlying bedrock as well (Table 2).

Following the geological and geophysical factors, the building characteristics are considered to be the second most important factor influencing indoor radon levels. With respect to radon entry pathways, the surveyed houses of Niška Banja are similar in their general construction, as seen in the documented building characteristics. The only factor which allows a distinction into subgroups to be made is the floor level on which the living room and bedroom are located. It should be noted that the floor designation correlates with some other building characteristics and can be considered as a generalized factor. In about 66% of surveyed houses the rooms are located on the ground floor. From the analysis of all data it was found that the indoor radon concentration is significantly higher at the ground floor level ($p = 0.04$, test of homogeneity of variances $p = 0.9$) than at upper floor levels. Assuming the type of underlying bedrock to be the major factor affecting indoor radon levels, it is necessary to consider separately subgroups of houses located in alluvium and travertine areas. Since only few data were available for rooms on the first floor in houses of alluvium area (four rooms) the analysis was performed only for houses located in the travertine area. This analysis proved that the indoor radon concentration is significantly higher at ground floor than at upper floor levels, as shown in Fig. 4b ($p = 0.001$, test of homogeneity of variances $p = 0.8$). The arithmetic mean annual indoor radon concentration in 29 rooms located on ground floor of houses over the travertine area of Niška Banja reached 2100 Bq/m^3, which is a very high level.

The generalized factors, type of underlying rock and floor, represent the primary factors influencing the indoor radon level in Niška Banja. To assess the relationship of primary factors (categorical variables) with indoor radon (numerical variable) the correlation ratio was estimated as a measure of association. Using four subgroups of data (logarithms of values of indoor radon concentration) obtained after categorization by primary factors, the correlation ratio is 0.51. This value indicates quite a reasonable association with respect to measured values of indoor radon concentration. The efficiency of analysis of secondary factors (the influence of which is assumed to be weak) is restricted by the need of further division of the data set into categories involving primary and secondary factors. For example, the lifestyle and habits of occupants of the houses surveyed represent a group of such weak factors.

Despite the above difficulties, a quite interesting correlation between indoor radon levels and indoor smoking was found. From the analysis of the entire uncategorized data set the highest values were observed in houses of non-smokers while indoor radon concentration was found to decrease with the number of resident smokers, as shown in Fig. 4c ($p = 0.046$, test of homogeneity of variances $p = 0.3$). Additional categorization by bedrock type and floor permitted only a significant decrease of indoor radon concentration to be observed in smoker's homes for travertine bedrock and first floor subgroup, while the effect of indoor smoking on indoor radon can be seen in most subgroups (Table 5). Introducing the presence of a resident smoker as the secondary factor results only in a minor increase of the correlation ratio, up to 0.56. The effect of resident smokers on indoor radon can probably be associated with higher ventilation rates in homes whose occupants smoke. The effect of smoking intensity was also analyzed and found not to be significant.

Some indication on the influence of the heating system installed in the house on indoor radon was also obtained. After categorization by type of underlying rock and floor level the only subgroup which allowed reliable comparison were houses located on alluvium sediments and the ground floor (Fig. 4d). In this subgroup we found statistically significant evidence of higher indoor radon concentrations in houses with solid fuel heating in comparison with houses with

Table 5
Geometric means of indoor radon concentration, Bq/m^3 (number of rooms surveyed in parentheses)

Number of smokers	Floor	
	Ground	First
Underlying rock – alluvium sediments		
0	470 (10)	34 (1)[a]
1 or more	373 (25)	200 (1)[a]
Underlying rock – travertine		
0	1712 (19)	706 (10)
1 or more	1259 (9)	227 (10)

[a] Insufficient data for analysis.

central heating ($p = 0.04$, test of homogeneity of variances $p = 0.66$). In this case the higher indoor radon concentrations may be associated with the well-known stack-effect.

Analysis performed on other documented factors resulted in no association with indoor radon levels. The following factors appeared to be not significant: presence of an anteroom, chimney opening in room, façade material, floor type in room, forced ventilation, hydro-isolation, source of the tap water, surface treatment of interior wall, wall type in room, and year of building construction. The chance that some of the above-listed factors do influence the indoor radon cannot be excluded. However, the limited sample of rooms available in our survey prevented the formation of subgroups of adequate size for proper investigation.

4. Conclusions

(1) Based on the observed values of outdoor and indoor radon concentrations, the region of Niška Banja should be ranked as a high natural background radiation area. In the studied buildings with low natural ventilation, located over travertine areas, the annual average indoor radon concentration was found to reach very high levels, with maximum values exceeding 6 kBq/m^3. Based on considerations of radon exposure dose conversion factors (such as those of the ICRP and UNSCEAR) the estimated annual effective dose may exceed 50 mSv with a regional average value of about 30 mSv. Average outdoor radon concentration levels are also relatively high, whereas the measured gamma dose rates remain in the medium range (UNSCEAR, 2000).

(2) The main factor influencing indoor radon levels in Niška Banja appears to be underlying bedrock type. Highest indoor radon concentrations were found in houses located over the travertine area. Travertine rock formed in the presence of radium in thermal spring waters constitutes a strong source of radon and provides the necessary routes for radon entry. The level in the house at which living rooms are located (i.e. ground floor or higher) represents a generalized factor of the building construction characteristics. Higher values of indoor radon concentrations on the ground floor confirm the role of underlying soil as the primary source of radon entry. The type of underlying bedrock and floor level are the two factors which significantly affect the variability of indoor radon levels at Niška Banja. Thus our results confirm that the geological and geophysical factors and the building characteristics are primary factors which influence the level of indoor radon concentration.

(3) Analysis of secondary influencing factors showed that higher levels of indoor radon concentration are observed in the homes of non-smokers. We explain this by more frequent

ventilation and higher indoor air exchange rates in the homes where smokers reside. The observation that indoor radon levels correlate with indoor smoking may be important for radon epidemiology (Darby et al., 2006).

(4) Our analysis of factors which influence the level of indoor radon concentration suggests some general principles for radon mitigation in the region. Indoor radon mitigation measures have to be directed towards preventing radon entry through the soil–basement–living room system. This could entail isolation of basement and living room floors. In some cases installation of a suitably arranged system of natural ventilation may be a cheaper approach. More costly measures, such as depressurization of basement space could also be considered. However, taking into account the building construction traditions of the region, designating living rooms to the upper floors and installation of central heating in newly erected buildings should be encouraged as the most readily available means of reducing the exposure of the citizens of Niška Banja to indoor radon.

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PAPER VIII

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Comparison of retrospective and contemporary indoor radon measurements in a high-radon area of Serbia

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Abstract

In Niška Banja, Serbia, which is a high-radon area, a comparison was made between two retrospective radon measuring methods and contemporary radon measurements. The two retrospective methods derive the radon concentrations that occurred in dwellings over longer periods in the past, based on the amount of trapped ^{210}Po on the surface of glass objects (surface traps, ST) or in the bulk of porous materials (volume traps, VT). Both surface implanted ^{210}Po in glass objects and contemporary radon in air were measured in 46 rooms, distributed in 32 houses of this radon spa-town, using a dual alpha track detector configuration (CR-39 and LR115) and CR-39 track etched detectors, respectively. In addition to the use of surface trap measurements, in 18 rooms (distributed in 15 houses) VT samples of suitable material were also collected, allowing to compare ST and VT retrospective radon concentration estimates. For each room, contemporary annual radon concentrations (CONT) were measured or estimated using seasonal correction factors. The distribution of the radon concentration in all data sets was found to be close to lognormal (Chi-square test > 0.05). Geometric means (GM) are similar, ranging from 1040 to 1380 Bq m^{-3}, whereas geometric standard deviations (GSD) for both the retrospective methods are greater than for the CONT method, showing reasonable agreement between VT, ST and CONT measurements. A regression analysis, with respect to the lognormal distribution of each data set, shows that for VT–ST the correlation coefficient r is 0.85, for VT–CONT r is 0.82 and for ST–CONT r is 0.73. Comparison of retrospective and contemporary radon concentrations with regard to supposed long-term indoor radon changes further supports the principal agreement between the retrospective and conventional methods.

Keywords: Indoor radon; Retrospective measurements; Long-term change

1. Introduction

For assessing the radon-related health risks, the need for reliable long-term radon data is obvious. The

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retrospective methods overcome the inability of the CONT methods to take into account past changes in radon exposure, for example in homes previously inaccessible to radon testing or subject to temporal and spatial variations in radon concentration (Ross, 2002). In recent years two methods of retrospective radon assessment have been developed, both based on measuring the activity of the long-lived radon descendant ^{210}Pb by means of its alpha emitting daughter ^{210}Po. The ^{210}Pb has accumulated over many years on the surface of solid media (mostly glass), known as surface traps (ST) (Samuelsson, 1988; Samuelsson et al., 1992; Lively and Steck, 1993; Falk et al., 1996) or in the bulk of porous media (mostly furniture filling sponges), known as volume traps (Oberstedt and Vanmarcke, 1996). Both methods are in fact based on the build up of low levels of ^{210}Pb and consequently ^{210}Po activity that can be related to the long-term average radon concentration. While the ST technique is based on solid state detectors (CR-39 and LR115), the VT technique uses radio-chemical procedures and alpha spectrometry. Both retrospective techniques have been used in surveys in the past in Serbia and elsewhere (Zunic et al., 1999, Paridaens et al., 2000, Birovljev et al., 2001) and a study by Paridaens et al. (2001) found the combination of VT and ST sampling to be very useful. Based on all previous field and laboratory work experiences as well as data analysis, in the present paper, a more detailed comparison of the ST technique devised in University College Dublin (UCD), Ireland and the VT technique devised by Oberstedt and Vanmarcke (1996) to CONT measurements, applied in the high-radon region of Niška Banja, Serbia is presented.

1. 1. Summary of radon studies in Niška Banja, Serbia

Niška Banja is a spa-town located 9 km east of Niš in Serbia (Fig. 1). The town is located in the quaternary alluvium of the river Nišava along the border of a karstic limestone region and thick strata of travertine (groundwater spring deposits). The formation of this travertine in conjunction with the high radium concentrations in the spring-waters of Niška Banja could be the cause of the high radium concentrations in the soils of the region. The measured radium concentrations in the soil are approximately 900 Bq kg^{-1} (Mazur et al., 2006). Due to the high radium content of the soil, a number of radon surveys have been conducted both indoors and outdoors (Zunic et al., 2006, Zunic et al., 2007) and the average indoor radon concentrations were found to be largely dependant upon the type of bedrock on which the dwellings were built. Dwellings built on the travertine plains were found to have, on average, radon concentrations of approximately 1550 Bq m^{-3} with some dwellings reaching 6000 Bq m^{-3}. For dwellings built upon alluvium sediments the average indoor radon concentrations was found to be 635 Bq m^{-3}, less than half that of those on the travertine deposits but still well above the Serbian national action level of 400 Bq m^{-3}. The average annual outdoor radon gas level was found to be 57 Bq m^{-3}, far higher than the typical worldwide value of 10 Bq m^{-3} (UNSCEAR, 2000). Due to the high values of indoor and outdoor radon concentrations, the Niška Banja region may be considered a high-radon area.

The majority of the indoor radon gas measurements during these surveys were contemporary radon gas measurements. However, in 2004 and 2005 a number of retrospective radon gas measurements were also conducted in various dwellings throughout this region. In a number of rooms, both ST and VT retrospective measurements were made, and here a comparison is made in these rooms where the two technique types were compared.

2. Materials and methods

2. 1. Contemporary radon measurements

CR-39 detectors, enclosed in small cylindrical (5 cm height, 3 cm diameter) diffusion chamber were used.

Fig. 1. Serbia.

The etched alpha tracks obtained counted with an automatic setup consisting of an optical microscope connected to a CCD camera controlled by personal computer. Exposure periods were generally of about 3 months covering one season. Annual averages were obtained using either results of all the seasonal measurements, if available, or results of some periods corrected with seasonal factors (Zunic et al., 2007).

2. 2. Surface traps technique

In the ST technique, short-lived airborne radon decay products deposit on smooth glass surfaces. Usually glass found in photograph and picture frames is used, because it is often easy to date simply by asking the owner about the age of the glass. After deposition, subsequent alpha decay can lead to implantation of further decay products through alpha particle recoil. Hence, the progeny can remain fixed in a very thin surface layer of the glass, where it can be detected through alpha decay by means of track etch detectors or pulse ionization chambers (Samuelsson, 1988; Samuelsson et al., 1992; Lively and Steck, 1993; Falk et al., 1996). A technique using CR-39 and LR115 detectors, the "CR-LR difference technique" aiming at large-scale fieldwork was used in the work described here to measure ^{210}Po (Falk et al., 1996).

2. 3. Volume traps technique

The VT technique (Oberstedt and Vanmarcke, 1996) is a trap technique, as it is also based on the accumulation of ^{210}Pb and subsequently ^{210}Po. However, it is the radon gas itself which diffuses rapidly and freely into bulky, porous materials, depositing there its decay products, thus these short-lived decay products are trapped in the VT and give rise to a build up of ^{210}Pb so that the radon concentration inside the VT is essentially representative of the radon concentration outside the VT in the room air. After about 1.5 years, the equilibrium between ^{210}Pb and ^{210}Po is reached and it is the latter isotope that has to be separated for analysis by radio-chemical means. The measurement material for the VT technique is typically a 100 cm^3 polyester foam sample taken from mattresses or cushions used in a dwelling.

However, it has been observed (Oberstedt and Vanmarcke, 1996) that spongy volume traps tend to accumulate 30 to 40% more ^{210}Po, than can be accounted for from pure volumetric considerations. This is due to the fact that some radon probably adsorbs to internal surfaces in the spongy volume trap much like it would on activated charcoal but to a far lesser extent. This effect has to be taken into account, and can be a source of possible error in estimating the retrospective radon concentrations. Therefore, the estimation of VT based retrospective radon concentrations in Niška Banja were corrected using the decreasing factor 0.65.

One of the major disadvantages is that the technique is destructive (i.e. the material is removed from the dwelling and destroyed during analysis unlike the surface trap technique) and so occupants are sometimes reluctant to supply samples for the measurement. In addition, it has been found that in very dusty circumstances direct penetration of radon progeny from the outside of a sample to the centre can be possible and so extra caution is needed in these circumstances.

2. 4. Placement and retrieval of detectors in dwellings

The fieldwork has been carried out by the members of the Institute of Nuclear Sciences Vinča (Belgrade). Surface trap detectors were installed in suitable rooms in the dwellings. A detailed questionnaire was completed for each room of interest that recorded a number of other relevant factors relating to the aerosol concentration, ventilation rate and surface to volume ratio in the past and present. These factors were then used in conjunction with the modified Jacobi room model (Jacobi, 1972) in order to better estimate the average radon concentration that the glass object was exposed to over its lifetime in the room.

In addition, during these investigations a number of these rooms had a sample of sponge material taken from the room for a volume trap analysis, the age of the material was also noted on a questionnaire. These samples were then sent to Belgium (SCK·CEN, Mol) for analysis.

The ST monitors were left in-situ for a period of approximately 3 months before being removed and sent back to UCD for analysis. Due to the extremely low intrinsic background of the transport detectors, the relatively short transport period in the post and the high ^{210}Po activity the detectors were exposed to, background and transport corrections did not influenced the results. The CR-39 and LR115 detectors were etched, counted and the retrospective estimate of radon concentration was obtained.

2. 5. Long-term changes of indoor radon concentration

When comparing the retrospective and contemporary measurements results it is necessary to take into account the long-term indoor radon changes (Yarmoshenko et al., 2005). Changes and transformations of construction components (such as basement, construction joints, insulation, interface gaps etc.), building underlying soil

Table 1
Parameters of distribution of indoor radon concentration estimated by three approaches

Measurements method	AM Bq m^{-3}	GM Bq m^{-3}	GSD	Percentage above 1000 Bq m^{-3}	Number of rooms
VT (all data)	2962	1121	4.3	53%	15
Contemporary matched to rooms with VT	1477	1067	2.5	53%	15
ST matched to rooms with VT	1475	1040	2.6	52%	15
ST (all data)	2728	1286	3.4	58%	46
Contemporary matched to rooms with ST	1956	1381	2.6	63%	46

physical condition and occupant living habits may in the course of time result in systematic changes of the radon entry. Analysis of retrospective indoor radon concentrations estimated over different ages of volume or surface traps can be somehow biased under condition of substantial long-term change of indoor radon. For example retrospective estimates of indoor radon concentration in the same space using two separate objects of different age may be found to be inconsistent. On the other hand the difference between radon concentrations determined by retrospective and contemporary technique can be rather explained by long-term changes than by strong measurements error. Thus the consideration of such processes allows meaningful comparison of contemporary and retrospective data as well as investigation of its pattern using these data.

By the results of radon entry modeling the long-term character of the indoor radon variation is close to linear (Yarmoshenko et al., 2005) though the year-by-year variation could be random. In particular, to describe monotonous long-term changes a coefficient k equal to the ratio of the annual radon concentration of two consecutive years has been introduced (Yarmoshenko et al., 2005). Using this coefficient, retrospective indoor radon concentration C_R (average of annual radon concentrations within a period equal to the age, A, of the "trap") is connected with contemporary indoor radon concentration C_C by equations:

$$C_R = \frac{1}{A}\sum_{i=1}^{A} C_C \cdot k^{i-1} = \frac{C_C}{A}\cdot\frac{k^A - 1}{(k-1)}$$

Using that approach coefficient k values are estimated for each pair VT–CONT and ST–CONT. Consideration of long-term indoor radon changes is meaningful on appropriate time scale. So for the following analysis we chose the 10 year period and estimated values of k^{10} (decade factor of radon concentration change). It was supposed that starting from this period the long-term indoor radon changes could prevail the random year-by-year variation.

Table 2
The results of the regression analysis (correlation coefficient and slope factor)

Pair (x–y)	Correlation coefficient, r	Slope factor (intercept=0)	N
VT–ST	0.85	1.13±0.50	15
CONT–VT	0.82	1.05±0.48	15
CONT–ST	0.73	0.93±0.23	46

3. Results

A total of 46 rooms (23 living and 23 bedrooms) in 32 houses of Niška Banja had ST measurements conducted in them. At least one surface monitor was placed in each dwelling. Fifteen rooms of 13 houses had VT measurements made in them.

The parameters of distribution of indoor radon concentration estimated by three approaches (CONT, ST, VT) are presented in Table 1. Distribution of radon concentration in all data sets is close to lognormal (Chi-square test, $p>0.05$), which is important for the following regression analysis.

The correlation of the three methods was studied using regression analysis of the log of radon concentration, due to the lognormal distribution of each data set, and it is presented in Table 2.

Further analyses involve to consider the indoor radon long-term change using the decade factors. Such

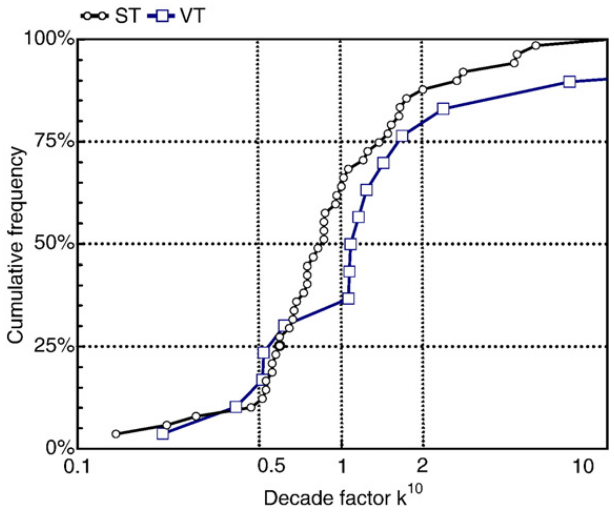

Fig. 2. Cumulative distribution of decade factors k^{10}. (ST: AM=1.5, median=0.87; VT: AM=1.48, median=1.09).

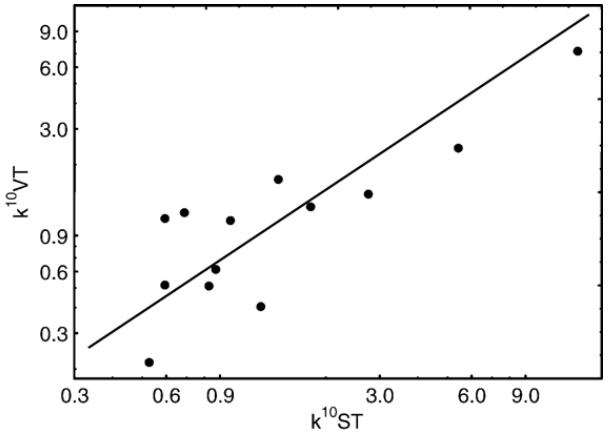

Fig. 3. Correlation of decade factors estimated using comparison of contemporary–VT and contemporary–ST approaches (equation of regression line $k^{10}_{VT}=(0.74\pm0.24)k^{10}_{ST}$, $r=0.8$).

approach allows to adjust the comparison for different ages of volume and surface traps. Cumulative distribution of decade factors k^{10} estimated using VT and ST methods are shown on Fig. 2. Substantial fraction (76%) of ST related results are in the reasonable range of k^{10}, i.e. from 0.5 to 2, which corresponds to maximum 2 times change of indoor radon concentration during 10 years. Most of ST based decade factors (61%) do not exceed 1, that corresponds to increasing pattern of long-term indoor radon changes. Fraction of VT related results within the range 0.5–2 is lower (67%). Contrary to ST, majority of VT based decade factors (67%) guesses long-term decrease of indoor radon concentration ($k^{10}>1$).

Fig. 3 shows the scatter plot of decade factors estimated using VT versus ST approaches. Among 15

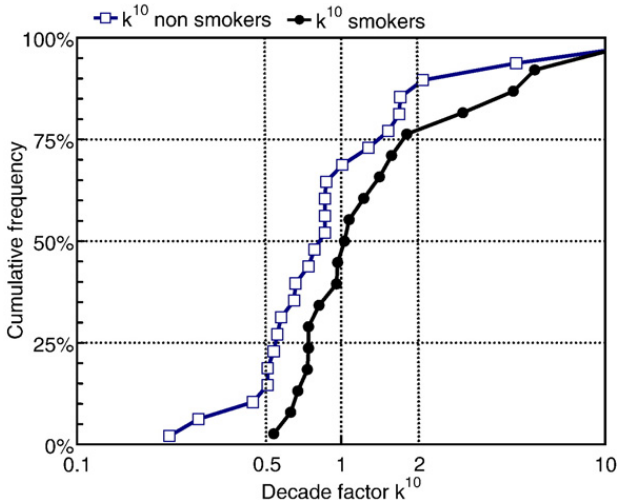

Fig. 4. Cumulative distribution of decade factors k^{10} estimated using ST retrospective measurements results separately for homes of non-smokers and smokers.

Table 3
Parameters of non-smokers and smokers houses

Parameter	Houses of non-smokers	Houses of smokers
Slope factor of contemporary–ST linear regression	0.84±0.27	1.20±0.46
k^{10} median value	0.83	1.03
Fraction of k^{10} ST<1	67%	47%
Fraction k^{10} ST in the range 0.5–2	75%	79%

pairs of decade factors obtained due to retrospective measurements in the same rooms 10 pairs represent the identical pattern of indoor radon long-term change.

Influence of different factors on relation between retrospective radon concentration measured by ST approach and contemporary measurements was investigated (amount of VT points is not enough for factor analysis). The following factors were considered: type of underlying rock, year of the construction, room type (living or bedroom), room floor, type of heating system, and tobacco smoking in house. Due to the results of the analysis only tobacco smoking is recognized to affect the relationship (24 and 19 rooms in houses of non-smokers and smokers respectively are available). Cumulative distributions of k^{10} for these two groups are presented in Fig. 4. Characteristics demonstrating difference for subgroups of houses of non-smokers and smokers are presented in Table 3.

4. Discussion

This study indicates good agreement between the two retrospective indoor radon estimation techniques and contemporary measurements. The GM and the parameters of GM and percentage above 1000 Bq m^{-3} presented in Table 1, show a reasonable agreement between ST, VT and contemporary measurements. Apparent differences in AM between retrospective and contemporary measurements follow from differences in GSD values. Higher GSD estimates for both ST and VT can be considered as evidence of higher measurements error of these methods in comparison with CR-39 diffusion type radon detectors. Highest average and GSD estimates were obtained for VT method. The satisfactory correspondence between ST, VT retrospective approaches and the contemporary CR-39 detector based method is also supported by the degree of correlation and the slope factor which is close to 1 (Table 2). Observed general agreement between VT based retrospective radon concentration corrected for factor 0.65 and both ST and contemporary measurements provides further evidence for adsorption of radon to sponge material or dissolution into it.

Consideration of long-term indoor radon changes using a comparison of ST and contemporary data further supports agreement between the retrospective and conventional methods. Estimated values of the decade factor that indicate indoor radon change during the past 10 years are quite reasonable considering known characteristics of the process. The relatively high degree of correlation between decade factors estimated using VT and ST methods (0.8) supports a conclusion on the coherence of these two retrospective approaches. At the same time according to significant deviation of the slope factor of linear regression (0.74 ± 0.24) from 1 ($p < 0.05$) complete agreement between the methods is not achieved. It means that VT and ST methods produce diverse assessment of indoor radon long-term changes. Such disagreement appeared for one third of the measurements. Also, it should not be forgotten, that if in certain dwellings strong disagreement exists between retro and CONT or between VT and ST results, this sometimes has obvious underlying physical causes. This was already shown in earlier comparisons (Paridaens et al., 2002) between retro and CONT results. One should check if such anomalies might perhaps be due to significant structural interventions in the dwelling or changes in the habits of its inhabitants, or other events at a certain point in the past. Also, the model of gradual long-term radon change might be more suitable in stable rural communities such as Niška Banja, than in areas with a more turbulent history such as mining areas or recently urbanized areas for example.

The observed dependence of relationship between retrospective and contemporary indoor radon concentrations on smoking in the houses can be caused by the following two reasons: 1) indoor radon long-term change may, through changes in ventilation practices, be influenced by smoking; 2) the "room model" developed to make conversions from ^{210}Pb surface concentration to radon gas concentration may not adequately take account of the influence of smoking generated aerosols on the ST technique. A similar effect of smoking, i.e. a lower correlation between retro and contemporary for houses with smokers smoking inside, was also observed in a previous study comparing ST and CONT radon concentration estimates in some Italian dwellings (Bochicchio et al., 2003).

The achieved results of a good agreement between retrospective and CR-39 contemporary method was assisted due to high indoor radon concentration level in houses of Niška Banja. This helped to ensure that the levels of ^{210}Po measured in both methods were sufficiently high for accurate measurement. The use of these retrospective methods in regions of low indoor radon level may be less reliable. Therefore, it is important for further studies that a protocol must be written when selecting volume and surface traps which are to be compared. This protocol would include comparisons in the exposure histories of each sample.

5. Conclusion

This study showed good agreement between the two retrospective radon gas estimation techniques and contemporary measurements. ST technique was much more accepted by householders than is the VT technique as the latter required pieces of furniture filling material to be destructively removed. Joint retrospective and contemporary indoor radon measurements, enhanced by the analysis of long-term indoor radon change, can improve exposure assessment reducing exposure uncertainties, which have a large impact on results of epidemiological studies.

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PAPER IX

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Radiation Measurements

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A campaign of discrete radon concentration measurements in soil of Niška Banja town, Serbia

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Abstract

The first radon soil gas survey in Serbia, using passive detectors (SSNTD, CR-39), was carried out in June 2005 at field sites in Niška Banja town. The aim of the survey was to identify risk zones characterised by high levels of this radioactive gas. Radon measurements were made at the depth of 50 cm, in the ground according to a systematic grid pattern. Furthermore, at all 48 measurement points, the surface gamma dose rates in the air was also measured at the same locations and soil samples were collected for gamma spectrometric analysis for the radionuclides ^{226}Ra, ^{228}Th and ^{40}K.

Radon concentrations were found to range from 1270 to 155 000 Bq m^{-3} with an average of 33 765 Bq m^{-3} and a median value of 12 626 Bq m^{-3}. The geometrical mean value and geometrical standard deviation were calculated as 16 160 Bq m^{-3} and 3.5 Bq m^{-3}, respectively. Gamma dose rate varies from 92 to 316 nGy h^{-1}, with an average of 132 nGy h^{-1}. The radium content in collected soil samples ranges from 24 to 1810 Bq kg^{-1} with an average of 187 Bq kg^{-1}. High correlations ($r^2 > 0.8$) between soil gas radon concentration, gamma dose rate and ^{226}Ra content in soil were found for each pair. The distribution of radon concentrations in soil gas shows bimodal shape.

Keywords: Soil gas radon; Radon spa; Travertine; Radium; High environmental radioactivity

1. Introduction

This paper deals with the results obtained during the first radon soil gas survey at Niška Banja town being inhabited with up to 6000 residents in its entire area and up to 15 000 ones spread out in surrounding villages. Niška Banja is a spa-town in the south-east part of Serbia, 10 km from the city of Niš and 224 km from Belgrade. The first indoor radon survey on more than 200 measurements in Niška Banja was conducted in the year 2000 using charcoal canisters (Manić et al., 2006) with reported radon concentrations up to 13 400 Bq m^{-3}. Following the request of the local community, further integrated long-term measurements of radon were made for all seasons of a calendar year in Niška Banja during 2003–2005 using CR-39 (SSI/NRPB) plastic alpha track detectors (Radosys). Indoor radon levels in many houses were found to be higher than the recommended value of 600 Bq m^{-3} and were as high as several thousand Bq m^{-3}, i.e., up to 6300 Bq m^{-3} (Žunić et al., 2006).

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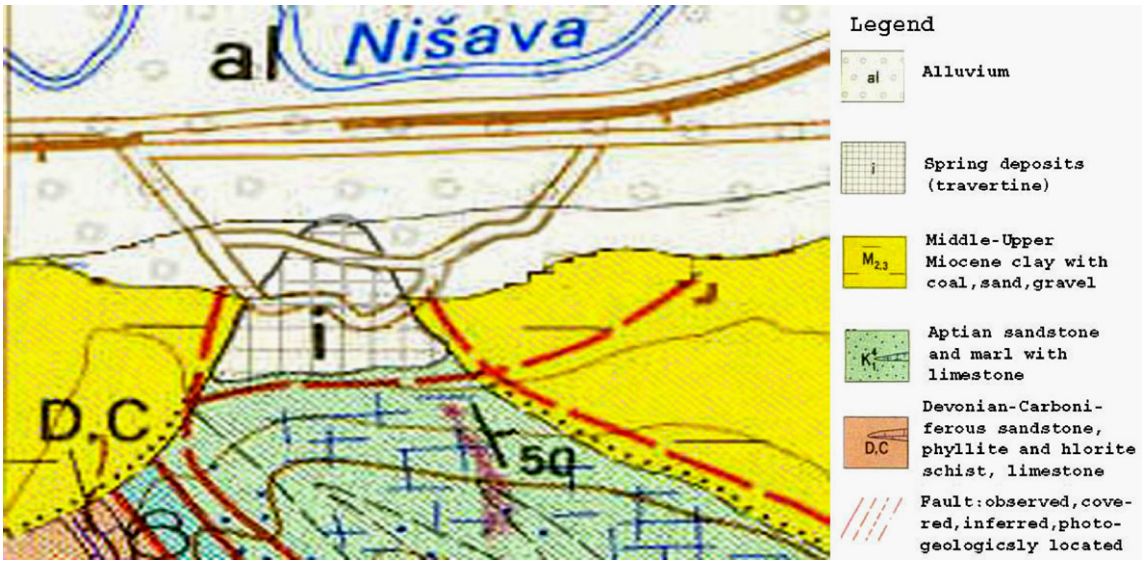

Fig. 1. Geological map of Niška Banja.

Outdoor radon concentrations were also recorded very high, up to 168 Bq m^{-3}, which is exceptionally high as an outdoor value (UNSCEAR, 2000). A significant correlation was found between high indoor radon concentration and travertine formation as its bedrock type (Žunić et al., 2007). Based on these facts and on previous results of radon measurements in soil gas (Žunić et al., 2006), the mapping of radon in soil gas of Niška Banja was found to be desirable and necessary for better understanding of the issue.

Due to radon short half-life, its diffusion in soil is limited at relatively short distances, so radon measured at the ground surface cannot be released from a deep origin, unless there exists a driving mechanism other than a mere diffusion. To explain radon migration over large distances, a deeper origin, however, is expected when ^{222}Rn of subsuperficial origin is carried upwards by a rising fluids, i.e., underground water or by carrier gases, such as CO_2, CH_4 , or N_2 (Kristianson and Malmqvist, 1982; Rogers and Nielson, 1991; Ciotoli et al., 1998; Etiope and Martinelli, 2002). These water and geo-gas discharges are strongly promoted in hydrothermal systems and seismically active zones.

From the geographical point of view, Niška Banja belongs to the mountain range known as the Carpathian–Balkan arch. It is located in the eastern part of Niš Neogene basin in the quaternary alluvium of the River Nišava along the border between Koritnik limestone and the thick travertine deposits. The formation of travertine in the presence of high radium concentration in the spring water of the Niška Banja region (Vučić and Pavlović, 1960; Vujisić et al., 1980) is similar to the well-known high natural radiation areas of Ramsar in Iran (Ghiassi-Nejad et al., 2002). According to this article, the high natural radiation in Ramsar is primarily due to the presence of very high amounts of ^{226}Ra in hot spring waters. Thus, the travertine enriched by radium may represent a source of radon and may allow appropriate migration routes through its porous structure.

The travertine plateau occupies the central part of Niška Banja town (Fig. 1) and its origin is of importance when the natural radioactivity of Niška Banja is considered. Travertine was formed in the quaternary period (Vujisić et al., 1980) as a result of carbonate precipitation from spring waters of Niška Banja. Travertine formation begins when water interacts with the soil zone, carbonate aquifers, organic material, or regional geothermal activity and produces a weak carbonic acid (H_2CO_3). This weak acid then reacts with limestone to form soluble calcium acid carbonate, $CaH_2(CO_3)$, which is precipitated when the water evaporates at some exposed surface, usually springs (Hurley et al., 1966; Friedman, 1970; Stumm and Morgan, 1981; Barnes et al., 1984; Cathelineau et al., 1989; Deines, 1992; Pentecost, 1995). The H_2CO_3 increases dissolution of carbonate rocks resulting in elevated dissolved CO_2 concentrations in the water. The concentration gradient between atmospheric CO_2 and dissolved CO_2 in water emerging from an aquifer initiates outgassing (Jacobson and Langmuir, 1970; Langmuir, 1971). As CO_2 concentrations move towards an equilibrium through outgassing, water becomes supersaturated with $CaCO_3$, eventually reaching a critical level. When this critical level is exceeded, a kinetic barrier is surpassed and $CaCO_3$ precipitates to form travertine deposits (Stumm and Morgan, 1981; Dandurand et al., 1982).

The mountain Koritnik is built up of the limestone (source of $CaCO_3$) and is one of the groundwater sources. Although there are reports about strong positive correlation between uranium and alkalinity of the water, expressed in mg/l of $CaCO_3$ (Ostle and Ball, 1973) this is no case of Niška Banja where disequilibrium between ^{238}U and ^{226}Ra is highlighted (Vučić and Pavlović, 1960). Since calcium and radium show a similar geochemical behaviour, the latter could replace calcium atoms in dissolved $Ca(HCO_3)$, forming $Ra(HCO_3)$ and then radium could precipitate on travertine mineral as $RaCO_3$. Radium

also can be directly precipitated on travertine by processes of adsorption or absorption.

The main objective of this paper is to integrate a variety of data including radon concentration in soil gas, radium content in soil and geological features, as well as gamma dose rate measurements in the air at a ground level to produce radon distribution maps which will be a useful input to health risk studies.

2. Experimental methods

In this study, SSNTDs were used for radon concentration measurements, employing Radosys type with CR-39. The measurements were performed over the entire town of Niška Banja in a grid pattern. Holes were drilled in the soil to the depth of about 50 cm depending on soil hardness. The CR-39 detector was placed at the bottom of the hole and then the hole was covered by a polyethylene bag, pressed with an appropriate stone (Fig. 2). Detectors were exposed for a period of 2 or 3 days only, due to the expected high radon concentration in the area. The CR-39 films were etched with the standard chemical method (20% NaOH, 90 °C, 4 h).

Gamma dose rates were measured at 1 m height above each hole using Environmental Meter type 6-80 and the measurements were performed with 5 min of averaging. GPS coordinates were noted for every measuring point and later were used for the mapping of soil radon concentration, radium content in soil and gamma dose rate values.

The soil samples were collected from each measuring point to determine the concentrations of natural radioactive elements—^{226}Ra, ^{228}Th and ^{40}K. They were taken from the depth of 20–50 cm. In the laboratory the soil samples were crushed and dried (105 °C) before putting into Marinelli beakers. After reaching the radioactive equilibrium they were measured using low-background spectrometer with $3'' \times 3''$ NaI(Tl) scintillation detector. The time of measurement was 30 000 s for each sample. The concentrations of natural radioisotopes were calculated using a "three-window" method. The IAEA materials (RGU-1, RGTh-1, and RGK-1) were used as standards.

Basing on the topographic map of Niška Banja town (scale 1:25 000) 48 measurement points were set and distributed in the 6×8 rectangular grid. The distance between two nearby points was about 250 m. Thus, the grid covered almost complete Niška Banja town area of 2×1.5 km (3 km^2), including all areas of interest from the geological point of view.

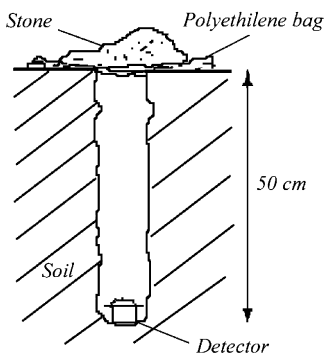

Fig. 2. Scheme of measuring point for radon in soil gas.

3. Results and discussion

GPS coordinates, soil gas radon concentration, concentration of natural radioactive elements in soil and gamma dose rate are given in Table 1. The soil gas radon concentration varies from 1270 to over a 155 000 Bq m^{-3}, with an average of 33 765 Bq m^{-3} and median of 12 626 Bq m^{-3}. The geometrical mean value and geometrical standard deviation were observed as 16 160 and 3.5 Bq m^{-3}, respectively.

The results of spectrometric analysis of soil samples, taken from holes, on ^{226}Ra, ^{228}Th and ^{40}K contents, show the range of radium content from 24 to 1810 Bq kg^{-1} (with an average of 187 Bq kg^{-1}), thorium content from 11 to 56 Bq kg^{-1} (with an average of 39 Bq kg^{-1}) and potassium content from 173 to 649 Bq kg^{-1} (with an average of 486 Bq kg^{-1}). Gamma dose rate varies from 92 to 316 nGy h^{-1}, with an average of 132 nGy h^{-1}. The results of the concentrations of radionuclides show the high level of ^{226}Ra content and non elevated content of ^{228}Th and ^{40}K (UNSCEAR, 2000).

The geographical distribution of radium content in soil is presented in Fig. 3. The similar distributions of gamma dose rate and radon gas concentration in soil are shown in Figs. 4 and 5, respectively. It can be observed that soil radon distribution (Fig. 5) corresponds to "Areas of high radon exposures—AHRE" (Manić et al., 2006).

The linear dependence between gamma dose rate in air and radionuclides (^{226}Ra, ^{228}Th and ^{40}K) content in soil bedrock is reported (UNSCEAR, 2000). Multiple regression analysis is the appropriate method for evaluation of the influence of several independent variables (^{226}Ra, ^{228}Th and ^{40}K contents in soil) on one dependent variable (gamma dose rate in air). Using the multiple regression analysis it was found that there is no statistically significant contribution to the gamma dose rate in air neither by thorium ($p < 0.36$) nor potassium ($p < 0.48$) contents in the soil, whereas the contribution of ^{226}Ra content in soil is highly significant ($p < 0.0001$). The correlation coefficients between different observed parameters are given in Table 2. The strong correlation is observed between gamma dose rate and radium content in soil ($r = 0.95$). It seems that the elevated gamma dose rate originates most probably from elevated radium content in the soil. Slightly less correlations between soil radon concentration and radium content in soil and between soil radon concentration and gamma dose rate, indicates the influence of other parameters such as soil permeability, ventilation of soil, water table level, etc.

Examining Figs. 3–5 it could be noticed that ^{226}Ra, ^{222}Rn and gamma dose rate values create anomalies which are approximately of the same shape for all three parameters. These anomalies are elongated structures in the form of a drop having a direction S–N. They are located along the water-course (formed from radioactive springs of Niška Banja) going from

Table 1
GPS coordinates, soil gas radon concentration, ^{226}Ra, ^{40}K and ^{228}Th concentrations in soil and gamma dose rate

Sample coordinate		^{222}Rn ($Bq\,m^{-3}$)	^{226}Ra ($Bq\,kg^{-1}$)	^{40}K ($Bq\,kg^{-1}$)	^{228}Th ($Bq\,kg^{-1}$)	Gamma dose rate ($nGy\,h^{-1}$)
N	E					
43°18′.106	21°99′.848	8542 ± 92	50 ± 2	494 ± 23	31 ± 1	116 ± 8
43°18′.129	22°00′.103	59 280 ± 240	278 ± 14	582 ± 31	42 ± 2	138 ± 8
43°18′.129	22°00′.311	42 990 ± 210	234 ± 11	623 ± 33	47 ± 2	149 ± 9
43°18′.081	22°00′.488	14 860 ± 120	69 ± 3	557 ± 26	39 ± 2	121 ± 8
43°18′.08	22°00′.691	49 770 ± 220	58 ± 3	446 ± 21	31 ± 1	102 ± 7
43°18′.092	22°00′.879	15 570 ± 120	35 ± 2	420 ± 20	29 ± 1	109 ± 7
43°18′.111	22°01′.068	9109 ± 95	62 ± 3	546 ± 26	40 ± 2	122 ± 8
43°18′.104	22°01′.257	7611 ± 87	59 ± 3	542 ± 26	39 ± 2	98 ± 7
43°17′.975	22°01′.281	10 160 ± 100	66 ± 3	649 ± 32	56 ± 3	140 ± 8
43°17′.964	22°01′.043	4310 ± 66	55 ± 3	495 ± 24	37 ± 2	121 ± 8
43°17′.997	22°00′.903	9919 ± 100	74 ± 4	604 ± 29	49 ± 2	122 ± 8
43°17′.986	22°00′.699	100 970 ± 320	532 ± 27	547 ± 34	44 ± 2	200 ± 10
43°17′.95	22°00′.485	> 155 000	1809 ± 110	318 ± 30	23 ± 1	316 ± 13
43°17′.975	22°00′.301	31 240 ± 180	150 ± 7	512 ± 26	36 ± 2	135 ± 8
43°17′.978	22°00′.167	24 900 ± 160	174 ± 8	626 ± 32	48 ± 2	124 ± 8
43°17′.97	21°99′.93	46 690 ± 220	249 ± 12	601 ± 32	43 ± 2	156 ± 9
43°17′.801	21°99′.972	24 800 ± 160	216 ± 10	607 ± 32	48 ± 2	138 ± 8
43°17′.834	22°00′.129	14 270 ± 120	116 ± 6	546 ± 27	42 ± 2	130 ± 8
43°17′.84	22°00′.318	13 260 ± 120	63 ± 3	518 ± 25	48 ± 2	129 ± 8
43°17′.778	22°00′.519	154 250 ± 390	1241 ± 69	297 ± 24	26 ± 1	251 ± 12
43°17′.824	22°00′.705	> 155 000	581 ± 30	369 ± 23	41 ± 2	209 ± 11
43°17′.827	22°00′.871	40 250 ± 200	204 ± 10	462 ± 24	33 ± 2	133 ± 8
43°17′.807	22°01′.081	12 410 ± 110	131 ± 6	562 ± 28	50 ± 2	142 ± 9
43°17′.811	22°01′.277	9413 ± 97	53 ± 3	573 ± 28	53 ± 3	138 ± 8
43°17′.703	22°01′.268	9311 ± 96	129 ± 6	260 ± 13	22 ± 1	115 ± 8
43°17′.698	22°01′.057	41 770 ± 200	222 ± 11	452 ± 24	43 ± 2	144 ± 9
43°17′.692	22°00′.859	22 860 ± 150	72 ± 3	485 ± 23	38 ± 2	116 ± 8
43°17′.694	22°00′.671	43 940 ± 210	174 ± 8	498 ± 26	46 ± 2	135 ± 8
43°17′.662	22°00′.533	> 155 000	781 ± 41	173 ± 12	11 ± 1	219 ± 11
43°17′.707	22°00′.331	49 520 ± 220	160 ± 8	399 ± 20	31 ± 2	112 ± 7
43°17′.693	22°00′.132	1273 ± 36	36 ± 2	487 ± 23	39 ± 2	121 ± 8
43°17′.697	21°99′.837	2630 ± 51	37 ± 2	584 ± 28	41 ± 2	93 ± 7
43°17′.584	22°00′.052	8379 ± 92	39 ± 2	601 ± 29	43 ± 2	120 ± 8
43°17′.571	22°00′.162	11 690 ± 110	41 ± 2	570 ± 27	39 ± 2	115 ± 8
43°17′.571	22°00′.311	10 290 ± 100	78 ± 4	378 ± 18	29 ± 1	114 ± 8
43°17′.556	22°00′.506	147 850 ± 380	126 ± 6	381 ± 19	32 ± 2	127 ± 8
43°17′.553	22°00′.689	12 840 ± 110	102 ± 5	434 ± 21	31 ± 2	111 ± 7
43°17′.568	22°00′.865	12 390 ± 110	40 ± 2	475 ± 23	41 ± 2	133 ± 8
43°17′.598	22°01′.061	25 540 ± 160	39 ± 2	480 ± 23	44 ± 2	105 ± 7
43°17′.583	22°01′.199	2468 ± 50	26 ± 1	318 ± 15	25 ± 1	92 ± 7
43°17′.498	22°01′.221	9290 ± 96	43 ± 2	502 ± 24	50 ± 2	100 ± 7
43°17′.49	22°01′.116	1576 ± 40	24 ± 1	385 ± 18	34 ± 2	97 ± 7
43°17′.455	22°00′.839	8500 ± 92	50 ± 2	482 ± 23	52 ± 3	93 ± 7
43°17′.478	22°00′.699	11 080 ± 110	55 ± 3	579 ± 28	55 ± 3	121 ± 8
43°17′.48	22°00′.521	5372 ± 73	30 ± 1	471 ± 22	42 ± 2	102 ± 7
43°17′.462	22°00′.407	2183 ± 47	51 ± 2	507 ± 24	49 ± 2	94 ± 7
43°17′.46	22°00′.185	2274 ± 48	36 ± 2	468 ± 22	43 ± 2	105 ± 7
43°17′.498	22°00′.023	8136 ± 90	29 ± 1	461 ± 22	29 ± 1	100 ± 7

hypsometrically higher to hypsometrically lower parts of Niška Banja, to the north, reaching river Nišava. The anomalies arise approximately at the place where thermal springs occur, then trace the water flow across the present travertine formation (probably creating younger travertine layers) and then entering the alluvial plane as progressing to the north, constantly spreading to the east and west until reaching its maximum at the stream curve (such places are common mineral deposition places). This leads to the conclusion that ^{226}Ra content in soil is directly connected to the water which introduces radioactivity to Niška Banja.

The concentration of soil gas radon does not follow the log-normal distribution (chi-square test, $p = 0.001$), this distribution seems to be bimodal. Thus, the measured values were divided into subgroups depending on the type of the underlying bedrock (travertine or alluvium sediments). In order to assign the study points to either travertine or alluvium sediments other investigated parameters can be used. Since the gamma dose

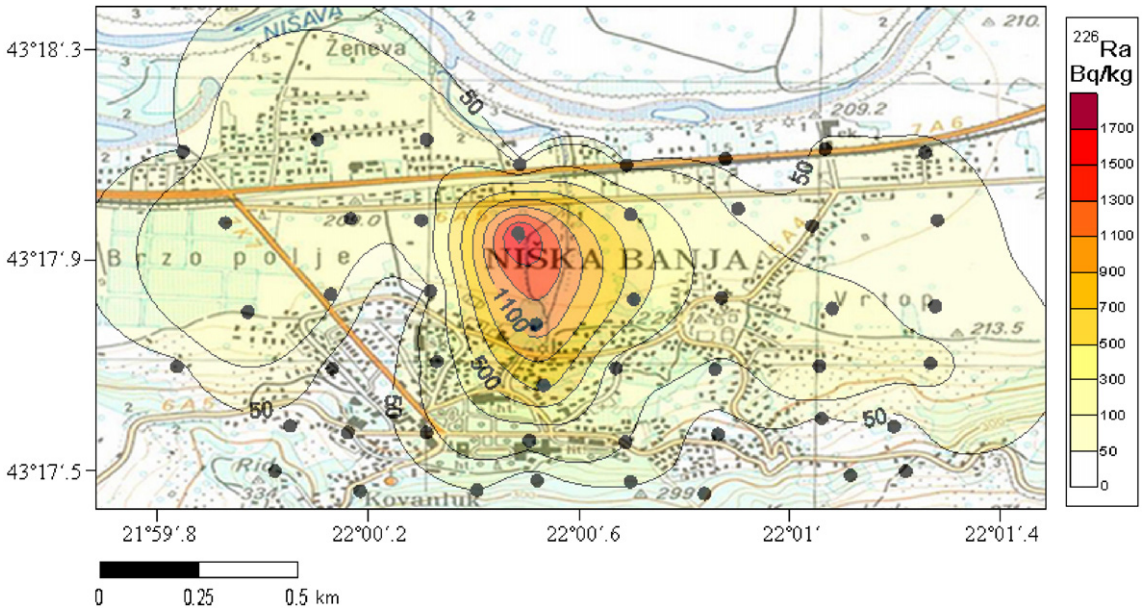

Fig. 3. Geographical distribution of ^{226}Ra content in the soil of Niška Banja (dotted—grid pattern of measurements points).

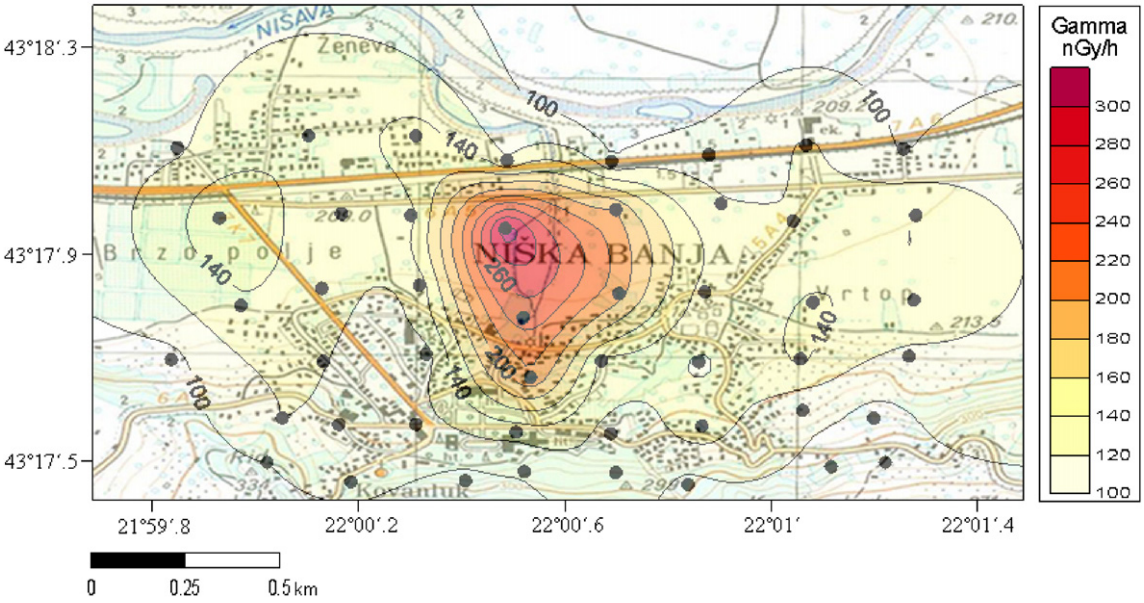

Fig. 4. Geographical distribution of gamma dose rate in the soil of Niška Banja (dotted—grid pattern of measurements points).

rate was measured at 1 m height above the holes it represents a reasonable generalised marker of soil radiation characteristics and can be used to delineate the travertine area. After the comparison of the geographical distribution of gamma dose rate (Fig. 4) with geological map of Niška Banja (Fig. 1) the value of 125 nGy h^{-1} was chosen to distinguish between travertine and alluvium sediment parts. Using this criterion the data set was separated into two subgroups. Data on ^{226}Ra in soil and soil gas radon concentrations were analysed in dependence on the location of measurement point in travertine or alluvium rock part of Niška Banja. The distributions of radium and radon in soil concentration are presented in Figs. 6 and 7, respectively. It can be seen that the distributions of the radionuclides over both travertine and alluvium sediment parts of Niška Banja are close to lognormal. The separation of the results due to the type of bedrock allowed to eliminate the deviation from lognormality in the initial dataset. The estimated arithmetic means (AM), geometric means (GM) and geometric standard deviations (GSD) are presented in Table 3. The average values of soil ^{226}Ra and ^{222}Rn concentration in travertine part exceeds these values in alluvium sediments part by factors 6 and 3.8, respectively.

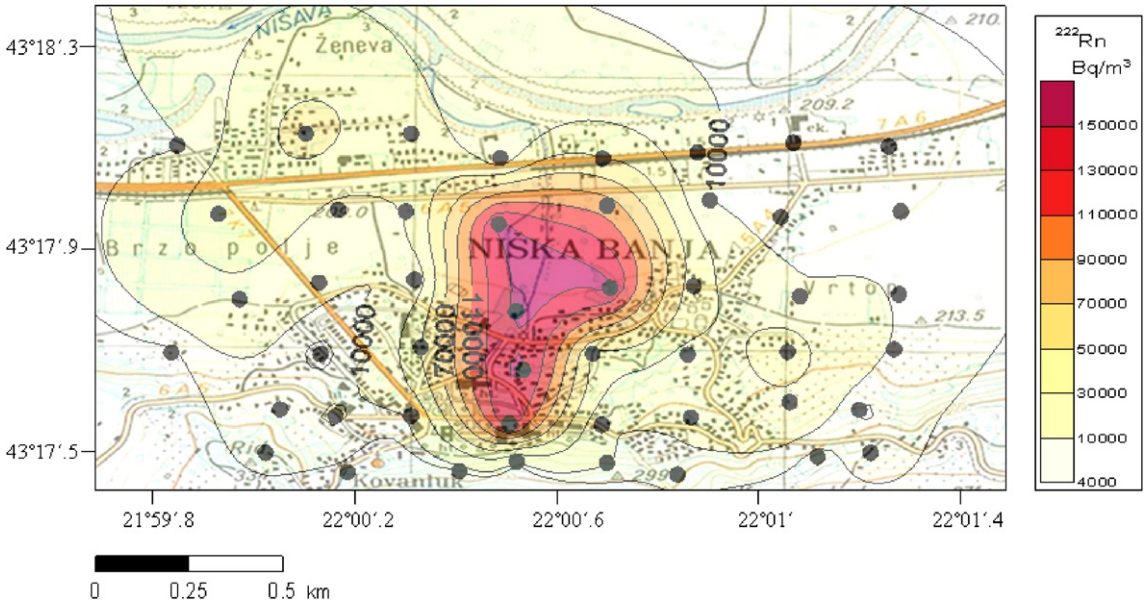

Fig. 5. Geographical distribution of radon gas (^{222}Rn) concentration in soil gas of Niška Banja (dotted—grid pattern of measurements points).

Table 2
Correlations between measured parameters

Measured parameters	Correlation coefficient r
Gamma dose rate–^{226}Ra concentration in soil	0.95
^{222}Rn concentration in soil gas–^{226}Ra concentration in soil	0.81
Gamma dose rate–^{222}Rn concentration in soil gas	0.82

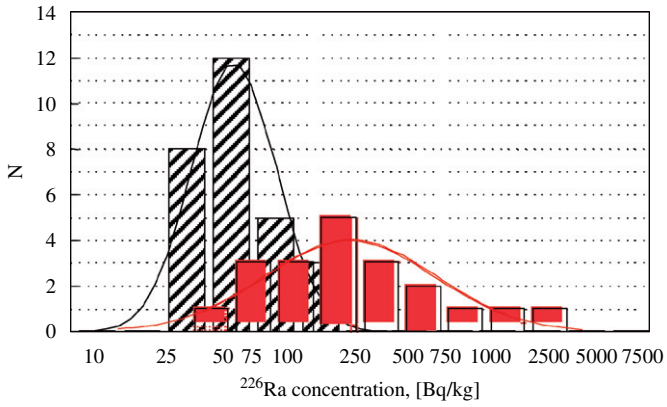

Fig. 6. Distributions of ^{226}Ra concentrations in soil for travertine (dark grey bars, solid line) and alluvium sediments (hatched bars, dotted line). Logarithmic concentration scale.

Based on the previous measurement results of the radon concentrations in soil (Žunić et al., 2006) and the measurements of indoor radon concentrations in Niška Banja during 2003–2005, it was reasonably assumed that high indoor and outdoor radon concentrations are connected with geology, especially travertine bedrock found in that region (Žunić et al., 2007). Although the main part of indoor radon originates from the soil gas, there is always an uncertainty in determination of the major source

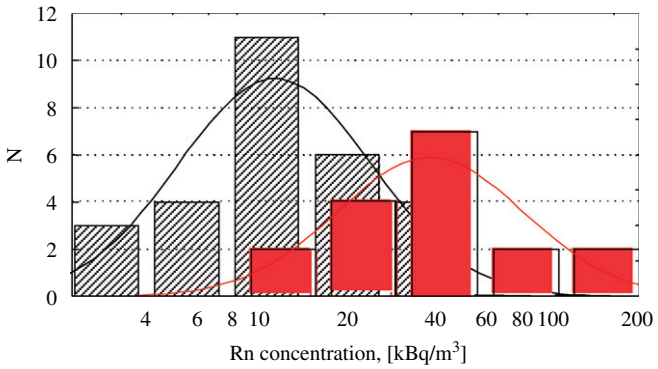

Fig. 7. Distributions of ^{222}Rn in soil concentrations for travertine (dark grey bars, solid line) and alluvium sediments (hatched bars, dotted line) parts of Niška Banja. Logarithmic concentration scale.

Table 3
Parameters of ^{226}Ra and ^{222}Rn in soil distributions

	Travertine bedrock	Alluvium bedrock
Distribution of ^{226}Ra in soil (Bq kg^{-1})		
Arithmetic mean	363	61
Geometric mean	213	53
Geometric standard deviation	2.8	1.7
Distribution of ^{222}Rn in soil (kBq m^{-3})		
Arithmetic mean	47.4	12.5
Geometric mean	32.2	8.3
Geometric standard deviation	2.5	2.6

of radon in the soil gas. Therefore, the knowledge and mapping of the radon concentration in soil gas could be an important method for the estimation of an area prone to elevated indoor radon concentration.

4. Conclusions

Based on the experimental results obtained in Niška Banja, the following conclusions can be drawn:

(1) When the soil is the relevant source of radon gas (as it seems to be in the case of Niška Banja, due to the presence of travertine as its geological background) it was necessary to perform a soil gas mapping for better understanding of the results of previous measurements. The values of radon in soil gas, gamma dose rate and ^{226}Ra content in soil of Niška Banja, show the significant correlations.
(2) Soil gas radon concentration at some places reached the values above 150 000 Bq m^{-3}. The values of radium content in soil and gamma dose rate at those points were found above 1200 Bq kg^{-1} and 250–300 nGy h^{-1}, respectively. Niška Banja is identified as the high level natural radiation area and thus should be more thoroughly examined, especially concerning epidemiological studies, biology, long-term immune and cytogenetic effects, etc. In addition, more data are required from the radon anomaly zone by considering the measurements in a denser grid, which is planned to carry out in the near future.
(3) The radon anomaly in Niška Banja is a local phenomenon even for aspects of a small town. Despite the fact that a small number of people live in this zone, this group should be the subject of systematic epidemiological studies, because of very high radiation doses they are subjected to. The advantage of this approach is that the control group for these studies could be chosen from other parts of the same town, where radon concentrations are at low levels. This is preferable because these people have the same habits and customs and the same genetic pool.
(4) It would be prudent to examine all houses situated in the radon anomaly zone as regards indoor radon and building techniques used. If houses with acceptable radon levels are found there this could be useful for proposing ways of radon mitigation. This approach to mitigation will introduce fewer disturbances among the people, which is very important considering the fact that people are very sensitive on any issue of radioactivity.

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PAPER X

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Radium-226 concentration in spring water sampled in high radon regions

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ABSTRACT

Water ^{226}Ra concentration in springs was measured in regions with high indoor radon: Ural, North Caucasus (Russia), Niska Banja (Serbia), Piestany (Slovakia), and Issyk-Kul (Kyrgyzstan). This paper presents the results for ^{226}Ra concentration above 0.03 Bq l^{-1}. Radium in water could indicate indoor radon problem in the region and water investigation is useful at the initial stage of radon survey. Even low ^{226}Ra concentration in water (0.1–0.6 Bq l^{-1}) caused high ^{226}Ra activity in travertine (up to 1500 Bq kg^{-1}), which resulted in indoor radon concentration above 2000 Bq m^{-3} (Niska Banja).

1. Introduction

For natural springs it is possible to find radium in water in a wide range of concentrations—from traces to significant values. Historically, most attention has been focused on ^{226}Ra and it is still the most important isotope. Radium is present in low-salinity solution as the uncomplexed cation Ra^{2+}. The weak complex of radium with chloride, sulphate and carbonate anions may be present in concentrated water, but will not affect the radium solubility or mobility to any extent in low-salinity water. It suggested that alpha recoil is a process for transferring radium from solid to aqueous phase. Once in solution, radium concentrations are controlled by sorbtion–desorbtion with the aquifer surfaces and, in water with high sulphate concentrations, by co-precipitation with insoluble sulphates (Dickson, 1990).

Radium in natural water springs presents two main problems in radiological protection. The first problem is the direct ^{226}Ra ingestion during water consumption. It is especially significant for spa resorts where the patients want to improve their health and pay more attention to the possible harmful factors. The second problem is concerning the fact that the elevated radium concentration in water springs can be an indicator of elevated ^{226}Ra concentration in deep geological formations or the source of possible soil contamination by ^{226}Ra and high radon concentration in the atmosphere of the dwellings situated over the contaminated place. Due to high radiotoxicity, ^{226}Ra is characterized by low action level (e.g. 0.5 Bq l^{-1} for drinking water in Russia) and a high-sensitivity technique is required.

2. Materials and methods

The modified method of ^{226}Ra concentration measurements is based on a well-known emanation technique (Lucas et al., 1990; Salih et al., 2000; Schubert et al., 2006): the specific activity of ^{226}Ra is determined by radon volume activity measurement in a sealed system during water sample bubbling. The advantage of this method is lack of any preliminary chemical or physical radium concentration and a small volume of sample (about 0.4 l). The measuring steps are as follows:

- degassing, sealing, and storage of water sample during at least 2 weeks in the flask, in which a sample was collected;
- absorption of background ^{222}Rn using the active coal filter cartridge in the inner volume of measuring system consisting of radon monitor AlphaGUARD (Genitron Instruments, Germany) based on pulse ionization chamber, set of glass vials AlphaKIT and air pump AlphaPUMP;
- transfer a water sample to a degassing vessel and air bubbling through the sample;
- measurement of ^{222}Rn concentration in a sealed system (at least 10 values of radon concentration for random error minimization);
- calculation of the specific activity of ^{226}Ra (A_m) by the following equation:

$$A_m = C\left(\frac{V_{\text{system}} - V_{\text{sample}}}{V_E} + k\right) - C_0\frac{V_{\text{system}} - V_{\text{sample}}}{V_{\text{sample}}} \quad (2.1)$$

where C– is the average radon concentration in the measuring system after expelling the radon, C_0– the background radon concentration in the measuring system, V_{system}– the inner volume of the measurement system, V_{sample}–volume of the water sample, and k– the radon distribution coefficient dependent on temperature.

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Table 1
Specific activities of ^{226}Ra and ^{222}Rn in water sampled in high radon regions and estimated effective doses relating to ^{226}Ra.

Country, region	Location	Description	Specific activity of ^{226}Ra ($Bq\,l^{-1}$)	Effective dose relating to ^{226}Ra ($mSv\,yr^{-1}$)	Specific activity of ^{222}Rn ($Bq\,l^{-1}$)
Russia, Ural	Lipovka (spa)	Radon spring	<0.03	–	540 ± 50
	Recreation Department Petushki	Drinking water	0.20 ± 0.03	4.0E−2	57 ± 6
	Recreation Department Razliv	Drinking water	0.35 ± 0.04	7.2E−2	92 ± 9
	Tyumen, Verkhniy Bor (spa)	Hot spring	0.21 ± 0.05	–	0.21 ± 0.05
Russia, Caucasus	Zheleznovodsk (spa)	Slavyanovskaya mineral water	4.1 ± 0.5	0.017	130 ± 15
	Zheleznovodsk (spa)	Smirnovskaya mineral water	2.1 ± 0.3	8.6E−3	118 ± 12
	Yessentuki (spa)	Yessentuki No. 4 mineral water	0.08 ± 0.03	3.3E−4	
	Yessentuki (spa)	Yessentuki No. 17 mineral water	0.05 ± 0.03	2.1E−4	
	Kislovodsk (spa)	Narzan dolomite mineral water	0.33 ± 0.06	1.4E−3	
	Pyatigorsk (spa)	Krasnoarmeyskiy mineral spring	0.09 ± 0.03	3.7E−4	
	Pyatigorsk (spa)	Mineral spring No. 2	0.36 ± 0.06	1.5E−3	
Kyrgyzstan, Issyk-Kul	Dzhety–Oguz spa	Drinking spring	0.97 ± 0.12	0.20	
	Dzhety–Oguz spa	Radon spring	1.8 ± 0.2	–	
Slovak Republic, Piestany	Spa, Napoleon Building	Mineral spring	1.8 ± 0.2	7.4E−3	56 ± 6
Serbia, Niska Banja	Skolska cesma	Drinking spring	0.11 ± 0.03	0.022	430 ± 46
	Kraljevo kupatilo	Hot spring	0.62 ± 0.10	–	44 ± 5
	Near Ozren Hotel	Hot spring	0.57 ± 0.09	–	148 ± 14
Serbia, other regions	Topilo spa	Drinking spring	0.08 ± 0.02	0.016	45 ± 5
	Kosovo, Gornja Stubla	Drinking spring	0.07 ± 0.02	0.014	
	Vranjska Banja	Hot spring	0.08 ± 0.02	–	
	Kosovo, Bozin bunar	Drinking spring	0.07 ± 0.02	0.014	
	Kosovo, Slatina mineralna	Drinking spring	0.11 ± 0.03	0.022	

Radon loss by decay during the measurement is neglected due to the short time of experiment. Special attention should be paid to background radon concentration and air-tightness of the system. To reach low detection limit in $0.03\,Bq\,l^{-1}$ the average value of background should be reduced to 5–10 $Bq\,m^{-3}$ and air-tightness of the system should be provided at the time of continuous bubbling of air through water. After 2–3 weeks' interval the water sample can be repeatedly measured in case of storage in the same completely filled flask.

The measurement error consists of systematic bias and random error. The systematic bias consists of AlphaGUARD calibration error (8%), error of the system interior volume assessment (1%), and error of sample volume (1%). The random error depends on a number of radon concentration measurements and a level of radon concentration in the system. The total error rarely exceeds 30%.

3. Results and discussion

The specific activity of ^{226}Ra in water was measured in five regions with high levels of radon in dwellings: Ural, Russia (Zhukovsky and Yarmoshenko, 1998); Niska Banja, Serbia (Zunic et al., 2007b); Piestany, Slovak Republic (Dubois, 2005); North Caucasus, Russia (Lezhnin et al., 2008); and Issyk-Kul, Kyrgyzstan (Zhukovsky and Termechikova, 2005). We have measured 55 water samples collected from different regions in Serbia, 24 samples collected from artesian springs in Issyk-Kul region, 9 samples taken from different spas of Caucasus, 14 samples collected from Ural springs, and one sample taken from a spa in Piestany. It was found that high radon concentration in water (400–600 $Bq\,l^{-1}$) does not always lead to the presence of radium in the sample and the relationship between ^{226}Ra and ^{222}Rn activity in spring cannot be estimated. In one case we obtain that all radon in sample is caused by ^{226}Ra decay, but usually ^{222}Rn activity in spring is higher than ^{226}Ra ones. Table 1 shows specific activities of ^{226}Ra and ^{222}Rn in samples having activities above low detection limit. In the majority of samples presented, the radium specific activity was either below action level or a little above it. The considerable excess of action level was obtained only in 4 water springs used for medical and bathing purposes.

The correlation between the presence of ^{226}Ra in water and high radon concentration in dwellings was found in Niska Banja and North Caucasus where the springs with radium content outflow for many years. For these places, the formation of travertine during the interaction of radium in water and limestone is typical. Fig. 1 shows that even low ^{226}Ra specific activity in water (0.1–0.6 $Bq\,l^{-1}$) in Niska Banja can cause high radium specific activity in travertine (up to 1500 $Bq\,kg^{-1}$) (Zunic et al., 2007a) and extremely high indoor radon concentration (more than 2000 $Bq\,m^{-3}$). For North Caucasus we did not find a direct spatial correlation between radium in surveyed springs and elevated indoor radon concentration (up to 1500 $Bq\,m^{-3}$), but the travertine formation with activity up to 1700 $Bq\,kg^{-1}$ (Tokarev and Shcherbakov, 1956) and a number of outflow springs in this region allow making an assumption concerning such correlation. For Piestany we cannot make any conclusions because of insufficient data. In Ural and Issyk-Kul region most springs are artesian with radium activity below 0.03 $Bq\,m^{-3}$ and formation of travertine does not occur.

To estimate committed effective doses via ingestion for ^{226}Ra, the considered adult dose coefficient was $2.8\times10^{-7}\,Sv\,Bq^{-1}$ (ICRP, 1996). Doses presented in Table 1 were calculated by taking

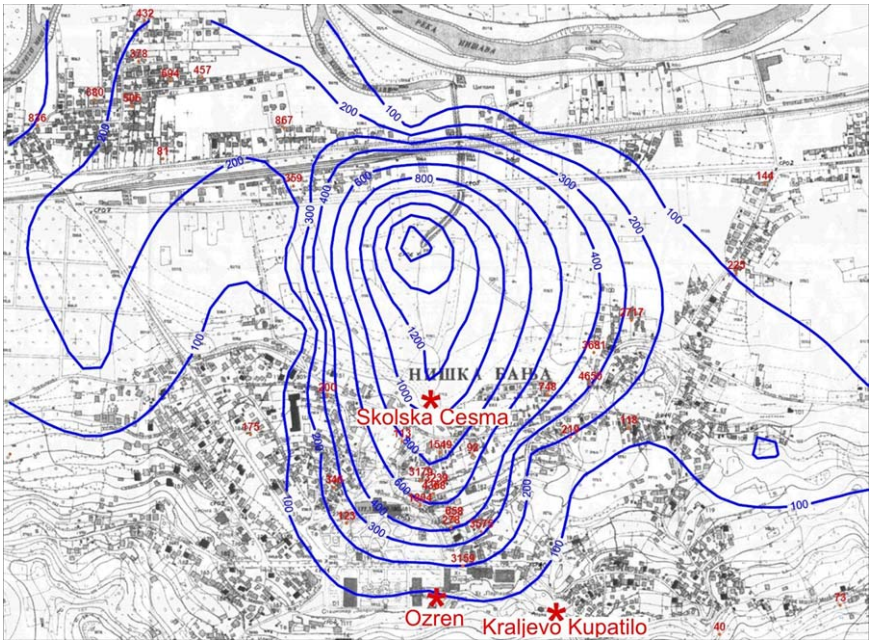

Fig. 1. Connection between the springs coordinates, isolines of ^{226}Ra concentrations in soil (Bq kg^{-1}) and average annual indoor ^{222}Rn concentrations (Bq m^{-3}) in Niska Banja, Serbia.

into account a consumption of 2 l of water per day for 1 year for drinking water and 0.7 l per day for 3 weeks for mineral water during cure process in spas. Doses from ^{226}Ra contained in hot springs and radon springs were not estimated due to another application—bathing. The action level of ^{226}Ra exceeded in a number of samples, but in general it was the mineral water for cure purposes with limited consumption; therefore, doses estimated for ingestion of radium in this waters were negligible. Only one case where the spring is used for regular drinking was found to exceed action level 0.1 mSv yr^{-1}.

4. Conclusions

In general, for the investigated regions there are no radiological protection problems due to radium consumption with drinking water. The elevated radium concentrations in water are typical for mineral water used in medical and recreation purposes. Nevertheless the presence of radium in springs can be an indicator of elevated indoor radon concentration in the dwellings of surrounding area and radium concentration measurements in springs could serve as one of the initial stages of radon survey.

Acknowledgments

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PAPER XI

Regular Paper

Field Experience with Soil Gas Mapping Using Japanese Passive Radon/Thoron Discriminative Detectors for Comparing High and Low Radiation Areas in Serbia (Balkan Region)

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Radon/Thoron/Natural radioactivity/Niška Banja/Obrenovac.

Based on results of fieldwork in the Balkan Region of Serbia from 2005 to 2007, soil gas radon and thoron concentrations as well as gamma dose rates were measured. Campaigns were conducted in two different geological regions: Niška Banja, considered a high natural radiation area, and Obrenovac around the TentB Thermal Power Plant (TPP), a low natural radiation area. Radon and thoron gas measurements were made by using two types of Japanese passive radon/thoron detectors, which included GPS data and gamma dose rates. The concentrations of soil radon gas in Niška Banja ranged from 1.8 to 161.1 kBq m^{-3}, whereas the concentrations for soil thoron gas ranged from 0.9 to 23.5 kBq m^{-3}. The gamma dose rates varied from 70 to 320 nGy h^{-1}. In the TentB area, radon concentration was found to range from 0.8 to 24.9 kBq m^{-3} and thoron from 0.6 to 1.9 kBq m^{-3}. The gamma dose rate ranged from 90 to 130 nGy h^{-1}. In addition, the natural radioactivity of the soil was investigated at the low background area. The radium and thorium contents in collected soil samples ranged from 23 to 58 and 33 to 67 Bq kg^{-1}, respectively. As a result of correlation analyses between the measured values, the highest correlation coefficient ($R > 0.95$) was found for thorium in the soil and the thoron gas concentration.

INTRODUCTION

According to a report by UNSCEAR[1)] the world mean annual effective radiation dose due to the inhalation of ^{222}Rn (radon), ^{220}Rn (thoron), and their decay products is estimated to be 1.26 mSv, with 1.15 mSv coming from ^{222}Rn and its progeny and 0.11 mSv from ^{220}Rn and its progeny.

In Niška Banja (located in the south-eastern part of Serbia; see the next section), several radiological surveys have been conducted. A preliminary (screening) survey was conducted for indoor radon/thoron.[2)] The soil-gas concentration ranges were from 63.7 to 1300 kBq m^{-3} and N.D. to 46 kBq m^{-3} for radon and thoron, respectively.

Niška Banja was identified as a high natural radiation area by this survey. Another detailed survey was conducted for radium in soil, radon in soil gas, and the gamma dose rate in a wider area of Niška Banja. The measurements show a significant correlation among radium in soil, radon in soil gas, and the gamma dose rate.[3)] Considering radon exposure and dose conversion factors such as that in ICRP 60[4)] and UNSCEAR, the estimated annual effective dose may exceed 50 mSv with a regional average of about 30 mSv.[5)] According to an earlier investigation, this value is close to the values for other high-level natural radioactivity areas (HLNRA).[6)]

However, the thoron concentration in soil gas was not

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measured in the whole area of Niška Banja, though previous studies have shown the existence of thoron in soil gas. Thus, the present study focused on soil thoron concentration in the Niška Banja area and its relationship between the gamma dose rate and soil radon concentration. As a control area (low natural radiation area), Obrenovac (TentB) (located in north-western Serbia, see the next section) was chosen.

In the Obrenovac (TentB) area, only a screening measurement was made and very low levels of radon and thoron concentration were reported.[7] Radioactivity concentrations of U, Th, and ^{40}K were also tested[8] for contamination of the soil and groundwater by thermal power plant waste, but no significant contamination was found.

The main objective of this paper is to compare the high and low background areas with a special attention to radon, thoron in soil gas, and their parent nuclide concentrations in soil, as well as the gamma dose rate measurement in the air at ground level.

Survey area

The present study was conducted at two places in Serbia. Niška Banja (180 km south-east of Belgrade) is regarded as an area with high-level natural radioactivity (HLNRA) and TentB (40 km south-west of Belgrade) as an area with low-level natural radioactivity (LLNRA).

a. Niška Banja

From a geological point of view, the town of Niška Banja is located in the quaternary alluvium of the River Nišava along the contact of Koritnik limestones and thick strata of travertine (spring sediments) end, which is also known as "bigar" in Serbian literature. This contact is also known as the "spa fault," which enabled the formation of a number of springs out of which the most important ones are Glavno Vrelo and Suva Banja.[9]

b. Obrenovac (TentB)

The lithology of the Nikola Tesla TPP (TentB) surroundings is very simple. It is built up from quaternary river sediments consisting of loam and argillaceous sandy sediments. These lithological members usually lie across river terrace sediments consisting of marl clay, carbonic clay, diatomaceous earth, and sands of the Lower Pliocene (Pontian). The sediments are concordant usually without any tectonic disturbances. The only significant tectonic structure (south of TentB) is a fault that stretches in the E-NE direction close to the villages of Dren and Grabovac. The geological and hydrogeological profiles of the Obrenovac area show the presence of a main sandy gravel aquifer between two impermeable formations of shallow alluvial clay-like sediments and Miocenic marly clay.[10]

MATERIALS AND METHODS

Measurements at Niška Banja were performed in November 2005 and at TentB in December 2006. Radon and thoron concentrations in soil gas were measured by using two types of passive radon-thoron discriminative detectors.[11,12] These detectors were developed and evaluated at the National Institute of Radiological Sciences (NIRS) in Japan.

They can measure both radon and thoron concentrations. One type of the detector, called RADOPOT, was deployed at 56 points in Niška Banja, whereas for measurements around TentB at 27 sites a new type of the detector called RADUET was used. This is a remodeled version of the RADOPOT monitor, which is now made in Hungary and is commercially available. A comparison of these detectors showed that the differences between the results do not exceed 10%.[13] Construction of these low cost detectors is simple, so the monitors have been widely used for radon and thoron surveys throughout the world.

This is the first time for RADOPOT and RADUET detectors to be used to measure radon and thoron concentrations in soil gas. The influence of humidity on radon/thoron measurement was tested at the NIRS and no significant influence (about 10%) was found.[14] The typical error on the radon/thoron concentration measurement was assumed to be 20–25%,[15] and therefore the humidity had only a small effect on the results.

Installation of the detectors at the survey areas was done as follows. Holes were drilled in the soil to a depth of about 80 cm depending on soil hardness. A detector was hung in the hole on a 70-cm stick and then the hole was covered by a polyethylene bag, which was pressed with an appropriate tile (Fig. 1).

Detectors were exposed for 4 days for Niška Banja and

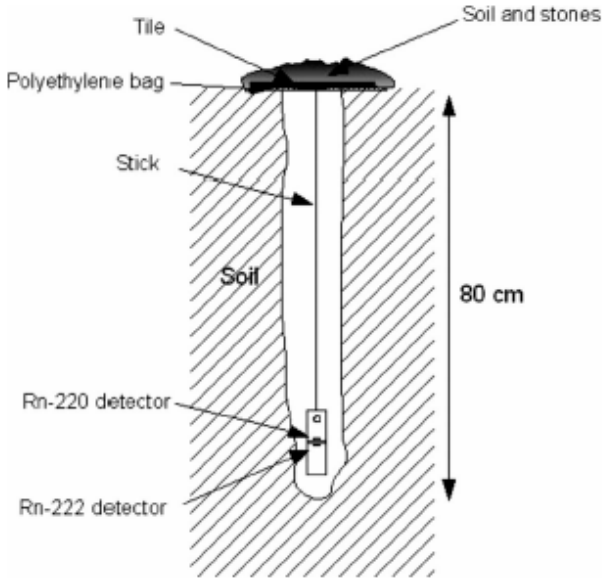


Fig. 1. Schematic of the measurement technique for radon/thoron in soil gas.

7 days for TentB, according to the expected radon concentration in the area. The CR-39 plates were etched for 6 h in 6.25 M NaOH solution at 90°C.[16)]

The measurements were performed in a grid pattern (with GPS coordinates) over the entire town of Niška Banja and around TentB, using a map of scale 1:25 000.

In the same locations, the gamma dose rate was measured using a scintillation counter. The measurements were performed 90–100 cm above ground level. Five parallel measurements (counting time: 100 s each) were conducted at every point. GPS coordinates were noted for every measuring point and later were used for mapping of the gamma dose rate and soil radon as well as the thoron concentration.

In addition, soil samples were collected from most of the measurement points in the TentB area to determine the concentrations of the natural radionuclides ^{40}K, ^{226}Ra, ^{228}Th, and ^{137}Cs. The samples were taken from a depth of about 50 cm and measured using a low-background NaI or HpGE detector. Samples were prepared in a plastic cylinder 9 cm in diameter and 4 cm high, closed, and aged for 2 to 3 weeks. The detector was characterized by Canberra, and an efficiency calibration made by using LABSOCS software (Canberra) was used for coincident summing corrections.

RESULTS

A total of 83 detectors were deployed for the measurement. Although detectors were deployed at 56 and 27 sites for Niška Banja and TentB, respectively, the concentration of soil thoron gas was not detectable at 6 sites. Moreover, data were excluded from further analysis if the measurement error exceeded the measurement value (16 cases). Results for concentrations of radon and thoron and for the gamma dose rates in both places are presented in Table 1. The Niška Banja to TentB ratios were calculated and are given as integers. The median value of radon concentration was 13.0 and 2.9 kBq m^{-3} in Niška Banja and TentB, respectively. The average radon concentration at each site was 26.5 ± 34 and 6.6 ± 7 kBq m^{-3}, respectively. The maximum radon concentration was 161.1 and 24.9 kBq m^{-3} with geometric mean of 14.5 and 3.9 kBq m^{-3}. The concentration of thoron gas was found with a geometric mean of 6.9 and 1.4 kBq m^{-3}, and ranged from 0.9–23.5 kBq m^{-3} for Niška Banja and 0.6–1.9 kBq m^{-3} for TentB.

The average radon concentration in the Niška Banja area was 4 times higher than around TentB and the thoron concentration was 7 or more times higher. The thoron/radon ratio for Niška Banja was more than 0.4 and 0.2 for TentB. The ratio of the gamma dose rate between the two areas was around 1.0. The average values were similar: 117 ± 50 and 114 ± 11 nGy h^{-1} for Niška Banja and TentB, respectively. The ranges of measurement values are different and amount to 250 nGy h^{-1} for Niška Banja with a maximum of 320 nGy h^{-1} and a minimum of 70 nGy h^{-1}, and a range of 40 nGy h^{-1} for TentB with a maximum of 130 nGy h^{-1} and a minimum of 90 nGy h^{-1}. These data suggest that the distribution of radioactivity in soil is heterogonous at Niška Banja (Figs. 2a, b, c) and homogenous for the TentB area (Figs. 2d, e, f).

As shown in the geographical distributions of the radon/gamma dose rate in Niška Banja, the area where the maximum value of radon concentration was found closely corresponds to the area of maximum gamma dose rate. In other cases, the distribution is flat. If the thoron concentration was

Table 1. Results for radon, thoron, and the gamma rate at Niška Banja and TentB.

Parameter		Area: Niška Banja (NB)	TentB (TB)	NB/TB ratio
Average + SD†	^{222}Rn [Bq m^{-3}]	26 500 ± 34 000	6 600 ± 7 000	4
	^{220}Rn [Bq m^{-3}]	10 600 ± 5 200	1 600 ± 500	7
	Gamma [nGy h^{-1}]	117 ± 50	114 ± 11	1
	^{226}Ra [Bq kg^{-1}]	–	48 ± 7	–
	^{228}Th [Bq kg^{-1}]	–	60 ± 8	–
Median	^{222}Rn [Bq m^{-3}]	13 000	2 900	4
	^{220}Rn [Bq m^{-3}]	8 400	1 700	5
	Gamma [nGy h^{-1}]	100	109	1
	^{226}Ra [Bq kg^{-1}]	–	49	–
	^{228}Th [Bq kg^{-1}]	–	62	–
GM (GSD)‡	^{222}Rn [Bq m^{-3}]	14 500 (400)	3 900 (3)	4
	^{220}Rn [Bq m^{-3}]	6 900 (200)	1 400 (2)	5
	Gamma [nGy h^{-1}]	–	113 (1)	–
	^{226}Ra [Bq kg^{-1}]	–	47 (1)	–
	^{228}Th [Bq kg^{-1}]	–	59 (1)	–
Range (min–max)	^{222}Rn [Bq m^{-3}]	1 800–161 100	800–24 900	–
	^{220}Rn [Bq m^{-3}]	900–23 500	600–1 900	–
	Gamma [nGy h^{-1}]	70–320	90–130	–
	^{226}Ra [Bq kg^{-1}]	–	23–58	–
	^{228}Th [Bq kg^{-1}]	–	33–67	–
No. of measurements	^{222}Rn [Bq m^{-3}]	56	22	–
	^{220}Rn [Bq m^{-3}]	49	5	–
	Gamma [nGy h^{-1}]	57	23	–
	^{226}Ra [Bq kg^{-1}]	–	23	–
	^{228}Th [Bq kg^{-1}]	–	23	–

†SD – standard deviation

‡GM – geometric mean and GSD – geometric standard deviations (in parentheses)

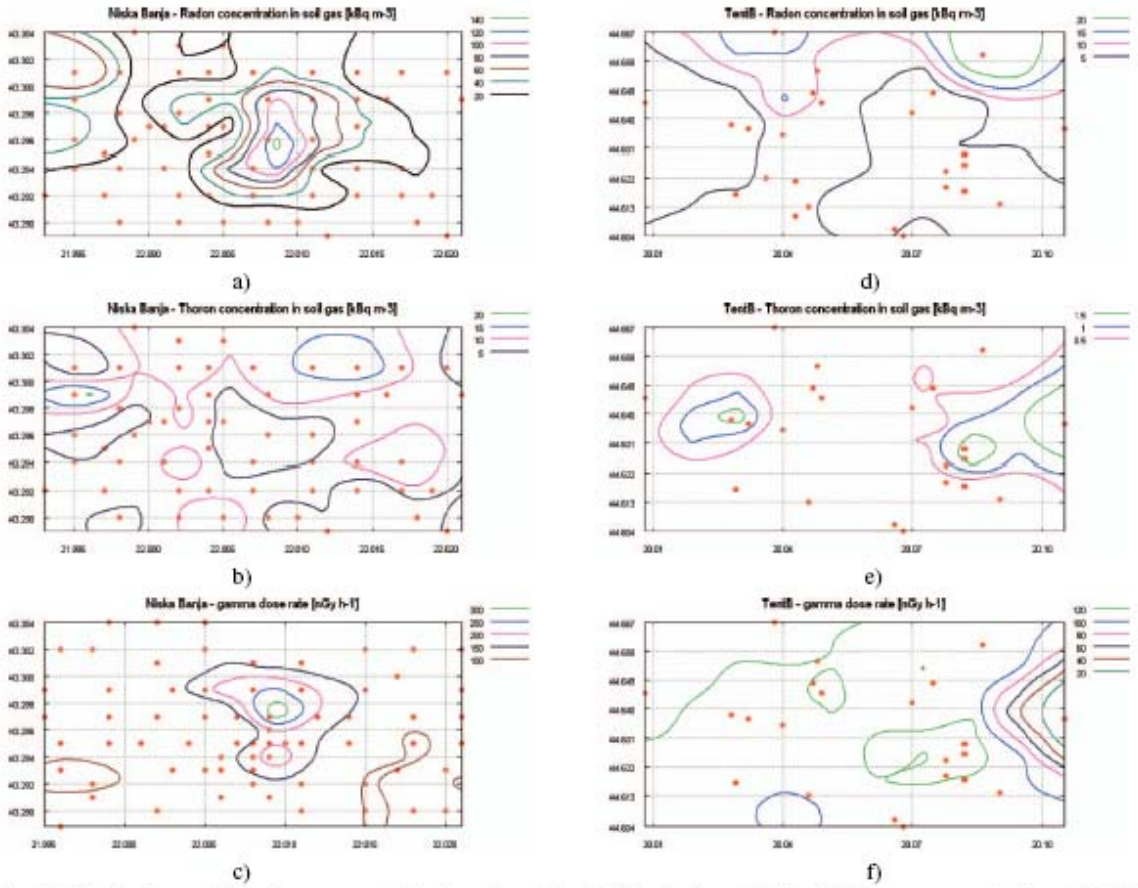


Fig. 2. Spatial distributions of the radon concentration in soil gas for a) Niška Banja and d) TentB, thoron concentrations at b) Niška Banja and e) TentB, and gamma dose rates at c) Niška Banja and f) TentB. The dots show the grid pattern of the measurements points.

lower than the error on its measurement, these data were not included in the spatial distribution of thoron for the TentB area (Fig. 2e). These data were also excluded from further analysis.

The averages of the gamma dose rate in both places were about 2 times higher than the world average (59 nGy h^{-1}). In neighboring countries UNSCEAR reported the following values: Slovenia, 56 nGy h^{-1}; Hungary, 61 nGy h^{-1}; Bulgaria, 70 nGy h^{-1}; and Romania, 54 nGy h^{-1}.

The soil samples were collected in the TentB area to determine the natural radioactive elements – ^{226}Ra, ^{228}Th, ^{40}K and ^{137}Cs. In a comparison between the Niška Banja and TentB results on soil thoron concentrations, it is interesting to analyze the data on ^{232}Th concentration in soil. The average ^{232}Th concentration in soil at TentB was 45 ± 6 Bq kg^{-1}, whereas a previous survey shows the average ^{232}Th concentration in Niška Banja to be nearly 40 Bq kg^{-1}. Nevertheless, the ^{220}Rn soil concentration in Niška Banja considerably exceeds the thoron soil concentration in the TentB region.

DISCUSSION

Natural radioactivity measurements were carried out at two areas in Serbia. The radon and thoron concentrations in soil in Niška Banja were higher than the concentrations found in soil in the TentB area, considered a "normal" background area. This can be explained by the differences in geological structure, content of natural radionuclides, and physical parameters of the soil.

Correlation analyses between measured values were performed and their results are presented in Table 2 for Niška Banja and Table 3 for TentB. By examining the Niška Banja results a strong correlation is observed between radon in soil and the gamma dose rate, with a correlation coefficient of 0.83. This result is close to the value obtained during an earlier investigation. Other calculated values presented in Table 2 shows no correlation (below 0.01).

An additional correlation analysis was done for TentB because this was the first investigation of natural radionuclides in soil. A spectrometric analysis for natural radionuclides in the soil samples resulted in average values of

Table 2. Correlation coefficients (R) between measured values for Niška Banja.

	^{222}Rn	Gamma	^{220}Rn
^{222}Rn in soil gas	1	–	–
Gamma dose rate	0.83	1	–
^{220}Rn in soil gas	< 0.01	< 0.01	1

Table 3. Correlation coefficients (R) between measured values for the TentB area.

	^{222}Rn	Gamma	^{226}Ra	^{228}Th	^{220}Rn
^{222}Rn in soil gas	1	–	–	–	–
Gamma dose rate	0.04	1	–	–	–
^{226}Ra in soil	–0.50	0.29	1	–	–
^{228}Th in soil	–0.31	0.31	0.82	1	–
^{220}Rn in soil gas	–0.27	–0.33	0.37	0.95	1

Table 4. Shapiro-Wilk normality test results.

Area / Parameter	Niška Banja		TentB	
	^{222}Rn	^{220}Rn	^{222}Rn	^{220}Rn
W	0.98	0.93	0.97	0.69
p	0.56	< 0.01	0.63	< 0.01

48 ± 7 Bq kg^{-1} (with a range from 23 to 58 Bq kg^{-1}) for radium and 60 ± 8 Bq kg^{-1} (with a range from 33 to 67 Bq kg^{-1}) for thorium. In the soil samples taken at a depth of 50 cm, an artificial isotope of ^{137}Cs occurred with an average concentration of 1.7 ± 1.8 Bq kg^{-1}. The source of this isotope is unknown and further investigation is needed.

The highest value of the correlation coefficient (R = 0.95) was obtained for the thoron and thorium analyses (Table 3). Also, the good correlation (R = 0.82) was obtained between ^{228}Th and ^{226}Ra. Assuming the secular equilibrium between ^{238}U series and ^{232}Th series and their progenies, the correlation between radon and thoron should be good, but actually not (R = –0.27). This may be because soil radon concentration is affected by other factors than ^{226}Ra concentration (correlation coefficient between ^{226}Ra and ^{222}Rn is –0.50).

In contrast to Niška Banja, for TentB the parameter R showed no correlation between radon and the gamma dose rate (R = 0.04). Other measured values also showed no close correlation with each other. Fig. 3a, b shows the correlations between the radon and thoron concentrations for both areas. Although the correlation coefficient was calculated, the analysis provided no correlation (or negative correlation) between these parameters.

The average ratio of thoron/radon for Niška Banja was close to 40%, whereas the range was very wide—from 1% up to 266%. At TentB, this ratio did not exceed 12% with a minimum value of 3%, so thoron could be neglected.

The Shapiro-Wilk test for normality was performed and the results are reported in Table 4.

The W-statistic and p-value were computed, from which a statistical decision can be made by comparison with a level of significance. The logarithm of the radon concentrations clearly followed a lognormal distribution (Fig. 4a–b), whereas in the thoron case the parameter "p - calculated probability" were below the border value (assumption significance level of 0.05) for both areas.

The simultaneous measurements of radon and thoron in soil gas at Niška Banja as well as at the TentB areas show that radon and thoron concentrations appear to be independent of each other. At some places in Niška Banja, the thoron concentration was 2 to 3 times higher than the radon concentration, whereas around TentB some places were found where thoron did not occur and the radon concentration was low (below 1.5 kBq m^{-3}).

Since the thoron problem has become worldwide, the

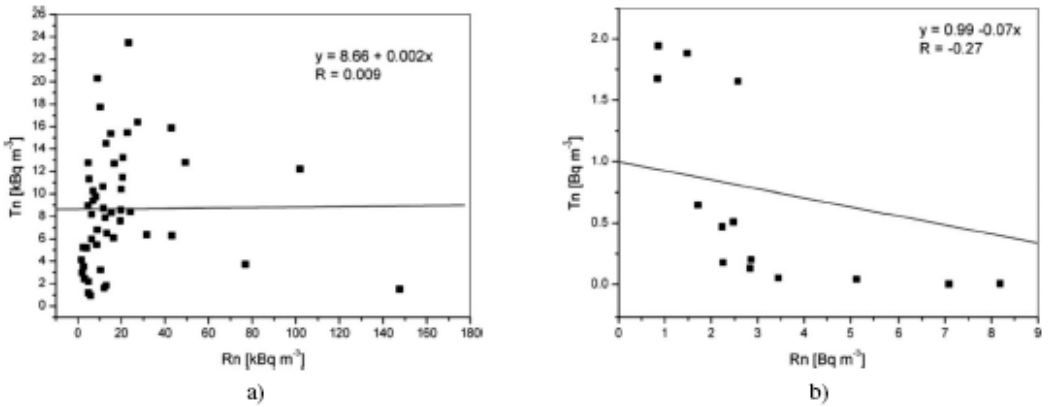
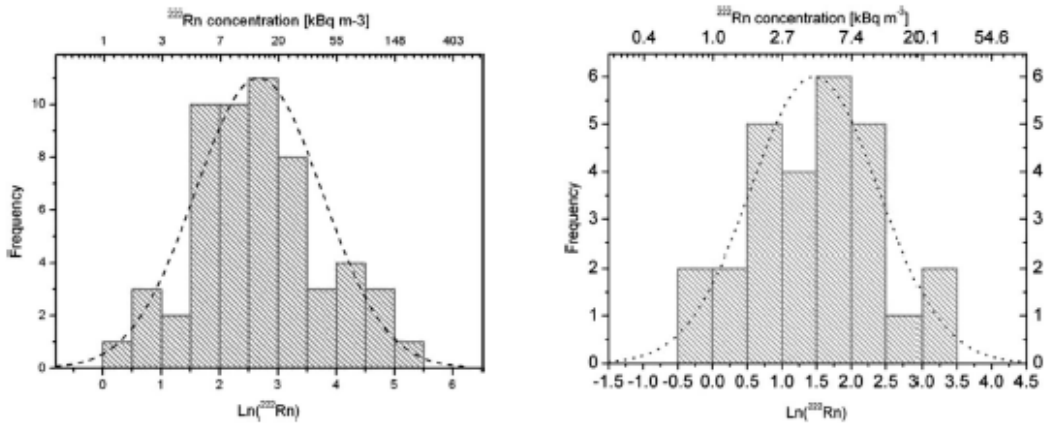


Fig. 3. Correlation between soil radon and thoron concentrations for a) Niška Banja and b) TentB.



a) b)

Fig. 4. Lognormal distributions for radon at a) Niška Banja and b) TentB.

importance of radon–thoron discriminative measurements should be recognized especially in areas with complicated geological structure and where high concentrations of radium or thorium are found.

To explain the difference between thoron and thorium in the soil at each site (at the same depth, the concentration of thorium in Niška Banja was lower than that at TentB while the concentration of thoron was higher) the physical parameters of the soil (such as permeability and diffusion coefficients) as well as the identification of mineral need to be determined and further study is required.

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B. A S S E S S M E N T

7 Assessment of the results

Radon concentrations were measured in many dwellings in the investigated areas. In many cases the concentrations were well above what are considered as unacceptably high levels from a health perspective for human exposure. For example in many countries there are ranges of radon action or reference levels above which it is recommended that remedial action should take place to reduce high radon exposures. These action or reference levels differ from country to country but are generally in the range 200 to 600 $Bq \cdot m^{-3}$ US EPA, 1987]. It is therefore necessary to estimate the lung cancer risk of the population in these communities in Serbia, where indoor radon levels exceeded the action or reference levels.

7.1 Health implications of long term radon exposure

Radon is one of a very small number of substances which have been confirmed to be human carcinogens based on human studies. It is considered Group 1 and Group A carcinogen as per the classifications used by the World Health Organization (WHO/IARC) and US Environmental Protection Agency [1987], respectively. The principal adverse health effect arising from the inhalation of radon and mainly its decay products is lung cancer. From a health perspective the most significant are radon (^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po) and thoron (^{216}Po, ^{212}Pb, ^{212}Bi, ^{212}Po) progeny. These elements, unlike radon, shortly after their formation attach themselves to aerosol particles, while a small fraction of them remain in unattached form, depending on aerosol size and concentration and ventilation rate [Marsh et al., 2005]. For the risk assessment of general population radon exposure (as distinct from occupational exposure) two different approaches can be used to estimate the lung cancer risk arising from exposure to radon decay products.

The dosimetric approach, in which the radiation doses to lung tissues is estimated and the associated risk is evaluated using currently accepted dose/risk factors for ionizing radiations and the residential epidemiology approach, which is based on the case-control studies of the general population are used to derive risk factors. In present study the risk to the exposed populations in the investigated areas of Serbia were estimated and compared using both approaches.

7.2 The dosimetric approach

In dosimetric approach, the dose to the lung is estimated using models of lung deposition of radon progeny and dosimetry. A number of such models exist and while they are similar in many regards they differ in a number of key assumptions such as: the identification and location of the sensitive cells in lung and the weighting factor for alpha radiation. In addition, at individual person level physiological factors such as lung morphometry, breathing rate, smoking habits, lung clearance rate, solubility of radon progeny in lung etc. would need to be taken into account. It is decided to estimate the doses to the general population, which includes both smokers and non-smokers, with the use of the value $65 \cdot 10^{-3}$ $mSv \cdot Bq^{-1}$ m^{-3} y^{-1} for the dose/exposure conversion factor for estimating the doses to the general population, which includes both smokers and non-smokers. It should be noted that the dose referred to is the "effective dose". This value is

derived from the work of Marsh, Birchall and Davis for equilibrium factor 0.4 and occupancy factor 0.8 [Marsh et al., 2005]. This value correspond to conversion factor $23 \cdot 10^{-6}$ $mSv \cdot Bq^{-1} m^{-3} h^{-1}$ from exposure on radon equivalent equilibrium concentration to effective dose. This value considerably exceeds the value $15 \cdot 10^{-6}$ $mSv \cdot Bq^{-1} m^{-3} h^{-1}$ estimated for dosimetric approach in UNSCEAR 2000 Report and the value $9 \cdot 10^{-6}$ $mSv \cdot Bq^{-1} m^{-3} h^{-1}$ recommended by the same report.

For estimating the dose and risk to the general population in the investigated areas the fatal cancer risk coefficient $5 \cdot 10^{-5}$ mSv^{-1} [ICRP21, 1991] is used for estimating the lifetime risk considering 70 years of lifetime. Estimated life time risk for radon induced lung cancer in 3 rural communities in Serbia: Kalna, Gornja Stubla, Niška Banja are estimated and presented in the table 7.1.

It is fully realized that a number of assumptions are made to derive these estimations. However, as they are not unreasonable these estimates may be taken as reasonable values at least for the purpose of comparing the radon risk between these three areas. It cannot be overemphasized that such estimates are not meant to apply at the level of an individual person but may be useful to decision makers and health authorities to develop policies and strategies for reducing future radon exposures and associated risks in these areas keeping in view the recommendations by ICRP and IAEA to reduce radon exposure in the houses.

7.3 The epidemiological approach

In past decades, epidemiological studies on the health effects of radon exposure have been made for underground miner cohorts (mainly on uranium and other hard rock miners). The results of these cohort studies have been extended to use for the estimation of risks to the general population exposed in their homes. As an example of such estimations the ICRP Publication 65 can be mentioned [ICRP23, 1993]. In this Publication the nominal mortality coefficient $3 \cdot 10^{-4}$ WLM^{-1} was recommended for radon progeny exposure both in working places and dwellings. It was supposed that this coefficient valid both to males and females in spite of the fact that only uranium miners data were analyzed.

In recent years, however, a number of major epidemiological case-control studies on the public (residential radon studies) have taken place, which gives more direct epidemiological information on the risk to the public. The detailed results of a collaborative analysis on individual data of 7148 persons with lung cancer and 14208 persons without lung cancer from 13 epidemiological studies in Europe has been published recently [Darby et al., 2006]. This is the largest and most comprehensive study of this type published in the literature to date. The excess relative risk of lung cancer per 100 $Bq \cdot m^{-3}$ was estimated to be 0.08 (95% CI 0.03-0.16) for raw data and 0.16 (95% CI 0.05-0.31) after adjusting confounding factors. The excess relative risk was not found to vary with age, sex or smoking habits. Obviously, the absolute risk of lung cancer for non-smokers is much lower than for smokers but the excess relative risk was essentially the same for both groups.

For assessment of radon induced lung cancer the official WHO statistic data presented on site www.who.int on total and lung cancer death rate in Serbia were used. According to these data the absolute lifetime risk to die from lung cancer in Serbia is 0.049 for males and 0.0069 for females. For assessment of lung cancer rate for non smokers the recommendations of ICRP Publication 50 [ICRP17, 1987] were used. According to these recommendations the lifetime lung cancer risk for non smokers at zero radon concentration was estimated at 0.0053 for males and 0.0037 for females. These estimations are in the good agreement with the risk estimation 0.0041 for the combine population made by Darby at al. [2006].

For the calculations of baseline lung cancer for smokers the 40% smoking prevalence for males and 30 % for females was assumed. According to the WHO data on total lung cancer death rate for population in Serbia and assessments of lung cancer rate for non smokers the absolute lifetime lung cancer risk 0.11 for smoking males and 0.035 for smoking females at zero or, more precisely, at average national radon concentration was obtained. Using the excess relative risk factor of 0.16 (given above) we can estimate the risk to smokers and non-smoker at 100 $Bq \cdot m^{-3}$ (for example) or any other radon concentrations in interest. It should noted that such simplified risk assessment approach when the total lung cancer risk calculated by multiplying baseline risk, risk coefficient and radon concentration is valid only for low and medium radon concentrations. For high radon concentrations, especially for smokers, the effect of risks competition should be taken into account.

The classical risk assessment with the use of Lubin [Lubin et al., 1994] and BEIR VI WL models [1999] was also conducted. On the basis of these calculations, assessments of the fatal lung cancer risk in the investigated areas: Kalna, Gornja Stubla, Niška Banja are presented in the Table 7.1. While the risks estimates using the dosimetric and epidemiological approaches are not in agreement with each other, they both indicate that the elevated radon in these communities may represent a significant health risk. It must also be stated and it cannot be overemphasized enough that all the above estimates (both using the dosimetric and epidemiological approaches) are based on many simplifying assumptions. Disregarding anecdotal non-scientific comment on lung cancer incidence in these communities, the actual values of lung cancer incidence in these communities as a function of age, sex, radon exposure, smoking habits etc. are not presently known. Nevertheless the risk estimates using either approach suggest that the high radon levels in these three communities (and in particular in Niška Banja) may represent a significant public health risk. From a positive perspective it is worth noting that building construction regulations now exist in a number of European countries. For new buildings, it possible to construct dwellings with low indoor radon levels even in areas where the rocks and soils contain elevated radon levels.

Table 7.1 Estimation of lifetime risk for lung cancer by dosimetric and epidemiological approach in 3 rural communities in Serbia

Parameter		Rural communities		
		Kalna	Gornja Stubla	Niška Banja
Average ^{222}Rn concentration, Bq·m^{-3}		188	447	1167
Average annual exposure by ^{222}Rn progeny, WLM·y^{-1}		0.84	2.0	5.2
Absolute radiogenic lung cancer risk assessment by dosimetric approach (whole population)				
Recommended UNSCEAR conversion coefficient		0.017	0.039	0.10
Estimated UNSCEAR dosimetric conversion coefficient		0.028	0.066	0.17
Marsh, Birchall and Davis conversion coefficient		0.043	0.10	0.26
Absolute radiogenic lung cancer risk assessment by epidemiological approach				
Assessment on the base of ICRP 65 Publication				
	(whole population)	0.018	0.042	0.11
Lubin model	males	0.042	0.096	0.23
	females	0.012	0.028	0.07
BEIR VI model	non smoking males	0.0053	0.018	0.046
	non smoking females	0.0037	0.012	0.030
	smoking males	0.10	0.22	0.45
	smoking females	0.035	0.081	0.19
Darby et al. model	males at whole	0.015	0.035	0.091
	females at whole	0.0021	0.0049	0.013
	non smoking males	0.0016	0.0038	0.0099
	non smoking females	0.0011	0.0027	0.0069
	smoking males	0.033	0.079	0.21
	smoking females	0.011	0.025	0.065

8 Conclusions

The indoor radon levels in dwellings of the investigated rural regions of the Balkans follow lognormal distribution (GSD=1.8-3.2). The recorded radon levels strongly depend on the type of underlying rock and the average radon levels were found to vary from 45 $Bq \cdot m^{-3}$ for limestone (Montenegro) to 1560 $Bq \cdot m^{-3}$ for travertine (Niška Banja).

The highest measured average indoor radon concentrations are about 1200 $Bq \cdot m^{-3}$ in Niška Banja, 500 $Bq \cdot m^{-3}$ in Gornja Stubla and 200 $Bq \cdot m^{-3}$ in Kalna. As estimated by parameters of lognormal distribution, part of houses with indoor radon concentration above 600 $Bq \cdot m^{-3}$ were 46% in Niška Banja, 23% in Gornja Stubla and 1.4% in Kalna.

There is the tendency of increasing indoor radon concentrations during the ageing of the dwellings, but retrospective radon measurements give the different values from expected value of decade increasing coefficient. Volume trap technique gives higher value of decade increasing coefficient than surface trap technique.

The season variations relating summer and winter indoor radon concentrations in Balkan region can be described by linear regression model ($C_{sum} = k \cdot C_{win} + A$) with the typical value of slope factor between the cold/worm season concentration k~0.2 - 0.6 (in dependence on seasons pairs) and intercept parameter A = 3 - 40 $Bq \cdot m^{-3}$. It is proved (Niška Banja) that there is negative dependency between the number of smokers in the dwelling and the average radon concentration and this fact should be taken into account during the epidemiological studies.

In some regions of Balkan (Gornja Stubla) the indoor thoron levels are significant and should be taken into account during both radon measurements and radiation dose and risk assessment.

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Appendix I Curriculum vitae

Full name: Žunić S. Zora

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Nationality: Serbian

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e-mail: zzunic@verat.net, ecelab@vinca.rs

Current appointment:

Researcher, Head of ECE Lab

Educational background:

1965 - 1973 Medical Faculty, University of Belgrade, **medical doctor**

1979, 1986 Control Department of Statens Serum Institute Copenhagen (UNDP/25), Radiation Microbiology, Specialization

1979 110th Basic Course of Radioactive Isotopes.

1997 Institute for Occupational Medicine Sector, Zagreb, Croatia, Specialization in radiation cytogenetics

2000 Medical Faculty, University of Belgrade, Master degree in experimental medicine course

Mr Sci Thesis Title: Population Exposure to Indoor Radon in Rural Communities

Professional background:

1993-1999 Head of Radiation Medicine Department while it was established: Radiation cytogenetic

laboratory, Laboratory for ant oxidative enzyme system and gave proposal for national radon survey in Yugoslavia

1999-2000 Organized and performed field work studies concerned with depleted uranium contamination in South Serbia within the project "Uranium in soil and plants of the Serbian Academy of Sciences"

2001 – within the first International Conference on Recovery of Yugoslavia (ENRY 2001), Belgrade organized depleted uranium session with eight platform presentation on experimental results from Southern Serbia (region Vranje, Kosovo and Metohija province, and Bosnia (Bratunac) and a field visit to region Vranje in South Serbia with a team comprising the scientists for Greece (NTUA), Poland (Institute of Nuclear Physics), Ireland (UCD) and UK (Middle Sax University)

2002 – Head of ECE Lab

2002/2003 Established Laboratory for Electro Chemical Etching(ECE LAB) being donated by NIRS, Chiba,Japan (Dr K.Fujimoto)

2003 – Initiated and Organized The first INTERNATIONAL ECE Workshop at

Vinca and Belgrade: "Promotion of New Electrochemical Etching Facility (ECE) and Its Application to Natural Radiation Studies in Western Balkan Countries", Belgrade, June 30 to July 3, 2003, Hotel Trim-Kosutnjak and Vinca Institute, ISBN 86-7306-060-5 Eds Z.S.Zunic, K.Fujimoto

2004-2005 - Head of Radiation Medicine Department

2005 – Initiated and Organized The second INTERNATIONAL ECE Workshop :

"The second Vinca ECE Lab advanced research international workshop – the new perspectives for thoron survey and dosimetry", Niška Banja, June 6 -10, 2005, Serbia and Montenegro, ISBN 86-7306-069-9, eds Z.S.Zunic, K.Fujimoto

2005 (October 22-26)– Organized and hosted as head of INTAILRISK and INDUWASTE projects (Serbia):

(1) the EU FP6 INTAILRISK Regular Project participant's meeting, SERBIA being project's participant, organizer

(2) the EU FP6 INDUWASTE Project participant's meeting and SERBIA being project's participant, organizer

(3) Field Intercomaparison Measurements within INTAILRISK Project request - field work at Niška Banja

Projects Involved:

1**.** ***EU FP6 - Programme***:

1.1 Assessment of Environmental Risk for Use of Radioactively Contaminated Industrial Tailings - Radioactivity of Coal and Bauxite in West Balkan Countries (acronym **INTAILRISK),** 1.1.2004 – 31.12.2007

1.2 Management and Remediation of Hazardous Industrial Waste in Western Balkan Countries (acronym **INDUWASTE),** June 2004 - 31.12.2006

2**.** Basic Research Project at the Ministry for Science and Environmental Protection of Republic Serbia: *Field Investigation on Population Exposure to Radon, Thoron and External Penetrating Radiation in Yugoslavia* , 2001- 31.12.2005

3. Bilateral State programs on Scientific Cooperation:

3.1 **Slovenia – Serbia**: Population Exposure in Slovenia and Serbia and Montenegro to Radon and Its Decay Products, Thoron and External Penetrating Radiation in its Human Residential and Occupational

Environmental Exposure, 2004 - 31.12.2005

3.2 **Greece – Serbia**: Relative Dosimetric Assessment On-Field and IN-the Laboratory Analysis of Risk from Depleted ad Natural Uranium 2005-2006

Agreement on Scientific Cooperation:

NIRS, Chiba, Japan, signed 2002

IFJ, Krakow, Poland, signed 2003

URORAN, Ekaterinburg, Russia signerd 2007

Current Scientific projects :

1 Nuclear Physics Methods and Instruments , P-141019 B: *Map of Radon Risk in Serbia* (Ministry of Science Republic of Serbia project period 2007-2010)

2 Technological project (Ministry of Science (2008-2009) *Genetic and radiation epidemiology regarding thermopower plant influence on population in its vicinity*

Appendix II Main Scientific Publications over the period 1999-2010

2010

Aleksandra Onishchenko, Michael Zhukovsky, Nenad Veselinovic, **Zora S. Zunic.** (2010) ***Radium-226 Concentration in Spring Water Sampled in High Radon Regions***. Applied Radiation and Isotopes, 2010, 68, 825–827

Gordana Milic, Bajram Jakupi, Shinji Tokonami, Radmila Trajkovic, Tetsuo Ishikawa,Igor Celikovic, Predrag Ujic, Olivera Cuknic, Ilia Yarmoshenko, Katica Kosanovic, Feriz Adrovic, Sarata K.Sahoo, Nenad Veselinovic, **Zora S.Zunic** (2010) ***The concentration and exposure doses of radon and thoron in residents of the rural areas of Kosovo and Metohija*** Radiation Measurements, 46 (1), 129-131

Predrag Ujic, Igor Celikovic, Aleksandar Kandic, Ivana Vukanac, Mirjana Djurasevic, Dusan Dragosavac, **Zora S.Zunic** (2009) ***Internal exposure from building materials exhaling ^{222}Rn and ^{220}Rn as compared to external exposure due to their natural radioactivity content***. Applied Radiation and Isotopes, Volume 68, Issue 1, January 2010, Pages 201-206

Zora S.Zunic, Nenad Veselinovic, Francesco Bochicchio, Vincio Carelli,Janja Vaupotic, Olivera Cuknic, Rodoljub Simovic, Zoran Vojinovic, Dragica Kisic, Tore Tollefsen (2009) ***The indoor radon survey in serbian schools: can it reflect also the general population exposure?*** Nukleonika Journal (submitted)

2009

Zunic ZS, Janik M, Tokonami S, Veselinovic N, Yarmoshenko I, Zhukovsky M, Ishikawa T, Ramola RC, Ciotoli G, Kozak K, Mazur J, Čeliković I, Ujić P, Onischenko A, Sahoo SK, Bochicchio F.(2009): ***Field experience with soil gas mapping using Japanese passive radon/thoron discriminative detectors for comparing high and low radiation areas in Serbia (Balkan Region).*** Journal of Radiation Research, 50, (4), 355-361.

Z.S.Zunic, I.V.Yarmoshenko, N.Veselinovic, M.V.Zhukovsky, P.Ujic, I.Celikovic, J.P.Mc Laughlin, S.E.Simopoulos, A.Birovljev, K.Fujimoto, J.Paridaens, F.Trotti, S.Tokonami, P.Olko, K.Kozak, F.Bochicchio, R.Ramola, J.W.Mietelski, B.Jakupi, G.Milic, G.Ciotoli, K.Kelleher, M.Budzanowski, S.K.Sahoo, H.Vanmarcke and M.P.R.Waligorski (2009), ***Identification and assessment of elevated exposure to natural radiation in Balkan region (Serbia),***Radioprotection,.44, (5), 919-925.

2008

Ujic Predrag, Celikovic Igo, Kandic Aleksandar, **Zunic ZS** . ***Standardization and difficulties of the thoron exhalation rate measurements using an accumulation chamber (Article)*** Radiation Measurements, (2008) vol.43 (8) 1396-1401.

Zunic ZS, Mietelski Jerzy W, Blazej Sylwia, Gaca Pawel, Tomankiewicz Ewa, Ujic Predrag, Celikovic Igor, Cuknic Olivera, Demajo Miroslav A. (2008): ***Traces of DU in samples of environmental bio-monitors (non-flowering plants, fungi) and soil from target sites of the Western Balkan region*****,** Journal of Environmental Radioactivity, (2008) vol.99 br.8 str. 1324-1328.

Vaupotic Janja, Celikovic Igor, Smrekar Natasa, **Zunic ZS,** Kobal Ivan: ***Concentrations of Rn-222 and Rn-220 in indoor air*****,** Acta Chimica Slovenica, (2008) vol.55 br.1 str. 160-165.

2007

Sahoo SK, Matsumoto M, Shiraishi K, Fujimoto K, Cuknic Olivera, **Zunic ZS**: ***Dose effect for South Serbians due to U-238 in natural drinking water*****,** Radiation Pprotection Dosimetry, (2007) vol.127 (1-4) 407-410

Zunic ZS, Yarmoshenko IV, Birovljev A, Bochicchio F, Quarto M, Obryk B, Paszkowski M , Celikovic I, Demajo A, Ujic P, Budzanowski M, Olko P, McLaughlin JP, Waligorski MPR : ***Radon survey in the high natural radiation region of Niška Banja, Serbia*****,** Journal of Environmental Radioactivity, (2007) vol.92 (3) 165-174

Zunic ZS, Yarmoshenko IV, Kelleher K, Paridaens J, McLaughlin JP, Celikovic I, Ujic P, Onischenko AD, Jovanovic S, Demajo A, Birovljev A, Bochicchio F: ***Comparison of retrospective and contemporary indoor radon measurements in a high-radon area of Serbia*****,** Science of Total Environment, (2007) vol.387 (1-3) 269 -275

Zunic ZS, Kozak K, Ciotoli G, Ramola RC, Kochowska E, Ujic P, Celikovic I, Mazur J, Janik M, Demajo A, Birovljev A, Bochicchio F, Yarmoshenko IV, Kryeziu D, Olko P : ***A campaign of discrete radon concentration measurements in soil of Niška Banja town, Serbia*****,** Radiation Measurements, (2007) vol.42 br.10 str. 1696 -1702

2006

Zunic ZS,, Kobal Ivan, Vaupotic J, Kozak K, Mazur J, Birovljev A, Janik M, Celikovic I, Ujic P, Demajo A, Krstic G, Jakupi B, Quarto M, Bochicchio F (2006): ***High natural radiation exposure in radon spa areas: a detailed field investigation in Niška Banja (Balkan region)*** **(Article)** Journal of Environmental Radioactivity, (2006) vol.89 (3) 249-260

2005

Zunic ZS, K.Fujimoto, I.V.Yarmoshenko**:** ***A Comparison of human exposure to natural radiation and DU in parts of the Balkan region***, International Congress Series 1276 (2005), 141-144

I.V.Yarmoshenko, **Zunic ZS,**, J.P.Mc Laughlin, J.Paridaens, I.A.Kirdin, K.Kelleher: ***Indoor radon long-term variation assessment,*** Radioactivity in the Environment, (A companion series to the Journal of Environmental radioactivity) eds:J.P.Mc Laughlin, S.E.Simopulos, F.Steinhausler, Volume 7 ISSN 1569-4860/DO1 10.1016/ S 1569-4860(04)07090-1,726 – 730, 2005

Gaca P., **Zunic ZS,**, J.W.Mietelski, E.Tomankiewicz, M.P.R.Waligorski**:** ***Experimental results on the environmental samples collected around sites in South Serbia, Kosovo and Montenegro where DU weapons were deployed in 1999,*** Radioactivity in the Environment, (A companion series to the Journal of Environmental radioactivity) eds:J.P.Mc Laughlin, S.E.Simopulos, F.Steinhausler, Volume 7 ISSN 1569-4860/DO1 10.1016/ S 1569-4860(04)07090-1, 1056 - 1063, 2005

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K.Sahoo, K.Fujimoto, K. Shirashi, **Z.S.Zunic**, ***Evidence of Deplated Uranium in South Serbian Environmental Samples***, Proceedings, 11th Interational Congeress of the Intenational Radiation

Protection Association, 23-28 May 2004, Madrid, Spain, CD Rom

S.K.Sahoo, K.Fujimoto, I.Celikovic, P.Ujic, **Zunic ZS,**, ***Distribution of Uranium, Thorium, and Isotopic Composition of Uranium in Soil Samples of South Serbia: Evidence of Depleted Uranium***, Nuclear Technology and Radiation Protection, XIX, 1, (2004), 26-30

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M. Esposito, P.Polic, B.Bartolomei, V.Benzi, M. Martellini, O.Cvetkovic, V.Damjanov, M.Simic, **Zunic ZS**,, B.Zivancevic, S.Simic, V.Jovanovic (2002): ***Survey of natural and anthropogenic radioactivity in environemntal samples from Yugoslavia***, Journal of Environmental Radioactivity 61, 271-282

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