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# IMPLEMENTATION OF THE EUROPEAN DIRECTIVE 96/29/EURATOM IN IRELAND – STATUS OF CURRENT INVESTIGATIONS

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The European Union Basic Safety Standard (BSS) Directive (96/29/EURATOM) is revised in line with scientific developments approximately every ten to fifteen years. The most recent revision, which took place in 1996, includes special provisions concerning exposure to natural sources of ionising radiation. Title VII of the revised Directive sets down a framework for controlling work activities where the presence of natural radiation sources leads to a significant increase in exposure to workers or members of the public, which cannot be disregarded from the radiation protection point of view.

The implementation of Title VII resulted in significant legal changes in Ireland. It has been incorporated into Irish law by a Ministerial Order (Statutory Instrument No. 125 of 2000) which came into force in May 2000.

This presentation will review the measures taken and progress achieved so far by the national regulatory agency, the Radiological Protection Institute of Ireland (RPII) since the coming into force of the new regulations in Ireland.

Based on their economical significance, a group of three major industries currently active in Ireland have been investigated from a list of work activities which, on the



basis of the literature, were considered liable to involve work practices resulting in exposure to NORMs. They include: the gas extraction and production industry, the peat-and coal-firing power generation industry.

For each of them, the RPII made an initial assessment of the scale of the problem by reviewing existing (non exhaustive) literature. Meetings with the industry respective management responsible for the personnel and/or the Health and Safety and/or the environment were organised to increase their level of awareness with regard to the current legislation and discuss of the potential consequences. This allowed us to compile information characteristic of each case and focus our interest on particular issues.

Field investigations were carried out and monitoring programmes put in place. Results obtained so far will be presented and preliminary conclusions will be drawn. Additionally, the bauxite/alumina refining industry (the largest European plant of this type is located in the West of Ireland), the cement industry and users of titanium oxide (pigment industry) and zircon sands are all scheduled to be investigated from 2004 onward. A brief overview of the radiological issues for each industry will also be presented.



# INVESTIGATION OF THE PEAT-FIRED POWER GENERATION IN IRELAND

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As the national agency with regulatory responsibility for ionising radiation in charge of the implementation of the most recent version of the BSS Directive 96/29/EURATOM, the Radiological Protection Institute of Ireland (RPII) initiated a programme in 2000 to identify types of industry currently active in Ireland, which on the basis of the literature were considered liable to (1) involve operations with and storage of materials not usually regarded as radioactive but which contain naturally occurring radionuclides causing a significant increase in the exposure of workers and, where appropriate, members of the public and/or (2) lead to the production of residues not usually regarded as radioactive but which contain naturally occurring radionuclides causing a significant increase in the exposure of workers and, where appropriate, members of the public.



Irish industries meeting one or two of the above conditions include the gas extraction industry, the fossil fuel power production (peat and coal-fired) and industrial processes using bulk materials with enhanced levels of NORMs (bauxite refinery, cement industry, mineral sands).

The Irish situation with regard to electricity generation is of particular interest as approximately 15% of the country's electricity requirement is provided through peat combustion. By 2004, only two large power plants will remain operative, representing a combined capacity of 250 MW. Between the two of them, just over 2 million tonnes of peat per annum will be used.

At the end of 2001, the RPII initiated a radiological investigation of the largest Irish peat-fired power plant in collaboration with Trinity College Dublin (TCD) and the Electricity Supply Board (ESB, the national Irish electricity supplier) to review the potential occupational exposures arising from the occurrence of NORMs at different stages of the industrial process. The objectives were to provide a valuable experience to the RPII in the frame of the new EU Directive, if necessary to provide the peat industry with advices and/or guidelines to adapt the production process and the environmental discharges to the new regulations, and finally to produce the first comprehensive radiological study of this industry.

Results of field investigations (external radiation dose rate measurements, aerial radon measurements, inhalation of airborne radioactive particles) and on-going monitoring of the radioactivity content of the peat, peat ash and effluent samples from the ash ponds will be presented as well as calculations of estimated radiation doses to workers involved in relevant work activities.





# **RADON EXHALATION RATE FROM SOIL USING FOR RADON MAP IN CLUJ CITY, ROMANIA**

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The majority of countries of the Carpathian Basin have no radon map. The goal of the paper is a proposal for a common sampling and measurement method to estimate the radon potential of this area.

The research was effectuated in 2003 summer and autumn period. The measurement points were chosen taking account the soil characteristics. Different soil types were investigated. The soil radon gas concentration was measured using track-etched detectors and the radon exhalation from soil was determined with accumulative method by ionization chamber.



In the same place we determined the radon exhalation from different depth respectively 20 cm, 40 cm, 60 cm, 80 cm. We also determined the radon soil gas concentration in these depths.

In the case of the accumulation method for the best fitting we need a measurement as long as possible. If the measurement time is too long the main problems are the back diffusion and the meteorological factors.

It is reasonable to use the longest measuring cycle because in this case the uncertainties of radon activity concentration measurements are the smallest.

The radon activity concentration values did not fit perfectly with the theoretical curve in case of soil concentration gradient method. Probably the reasons are the inhomogeneous and perturbed soil structure. We suggest exposure time 3–4 days depending on the depth of measurement points.



# **A COMPARATIVE ANALYSIS OF THE HEAVY METALS, HEAVY NATURAL RADIONUCLIDES CONCENTRATIONS AND BIOINDICATION ASSAY OF WATER SAMPLED FROM A RADIUM PRODUCTION INDUSTRY STORAGE CELL TERRITORY**

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Mining and processing industries are main sources of environmental pollutants: heavy metals, heavy natural radionuclides and others. This leads to many ecosystem undergoing a simultaneous potential stress from chemical and radioactive toxicants. Our previous works demonstrates that in order to achieve pollutant screening, it is not sufficient to determine the pollutants concentration only. The chemical analysis data reveals nothing about the total exposure an organism has received. In contrast to the specific nature of assessments on exposure, effect studies inte-



grate the impacts of all mutagenic activities, including synergistic and antagonistic effects. Consequently, adequate conclusions on the risk due to environment contamination need to be based on the additional simultaneous use of toxicity and genetic toxicity tests.

In this report results of the simultaneous use of bioindication and chemical analysis approaches for estimation of the chemical and radioactive contamination of the water from reservoirs located near the radium production industry storage cell are represented for the first time. The storage cell is situated in the settlement of Vodniy (Komi Republic, Russia). A plant for the production of radium from underwater had been in operation at this place from 1931 until 1956. In particular, in this report will represent results of:

comparative analysis of modern and observed in 1959–1960 levels of radioactive and chemical contamination of the water from reservoirs located near the radium production industry storage cell;

genetic toxicity and toxicity assay of water sampled from a radium production industry storage cell territory by means of Allium-test.

Further to this, the attempts were made to understand whether or not any correlation existed between heavy metals, heavy natural radionuclides concentrations and observed biological effects. In addition, possible mechanisms of genotoxic and toxic action of radioactive and chemical combined environmental contamination will discuss.



# A COMPARATIVE ANALYSIS OF BIOLOGICAL EFFECTS IN PLANTS INHABITING TWO RUSSIAN REGIONS WITH A HIGHER LEVEL OF HEAVY NATURAL RADIONUCLIDES

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Extracted on a ground surface rocks and reservoir water are contain heavy natural radionuclides (HNR), which both  $\alpha$ -,  $\beta$ -,  $\gamma$ -emitters and high-toxic chemical elements. As it is the case for man, the HNR seem to constitute a preponderant source of exposure to natural radiation for the majority of living wild organisms. Despite of rather high HNR levels in a refuse heaps, the problem on their possible effects on plants and animals inhabiting spoil heaps of mines was investigated only in a few studies.



In this report the data on a content of isotopes of U, Ra and Th in soils and plants of two northern region of Russia (Komi Republic (radium-bearing waste) and South Yakutia (uranium-containing waste pile)) are represented. Despite of similar character of a radioactive contamination and a close climatic conditions, the radiobiological effects in plants from these regions were drastically differing. The higher frequency of aberrant cells in *Vicia cracca* L. germs from Komi Republic site was found. Moreover, the elimination of bearing chlorophyll mutations plants was observed in these *Vicia cracca* L. populations. The seeds of *Vicia cracca* L. plants from Komi Republic site were significantly more sensitively to acute  $\gamma$ -radiation when compared with untreated control. In contrast, no significant difference in aberrant cells frequency of alder trees seedlings growing on the South Yakutia site was noted. In addition, the seeds from the alder trees populations growing in the South Yakutia site appeared to be significantly more resistant to the acute radiation exposure than the reference seeds.

The reason for these differences is not clear now. It is reasonable to assume that at least some of this variability can be accounted for by differences in radiation exposure structure. In Komi Republic site the contribution of the internal irradiation to an equivalent dose is 3–3.5 times above share of the external  $\gamma$ -irradiation. Unlike Komi Republic site, for South Yakutia site the external  $\gamma$ -irradiation (71–86%) play a decisive role. Future studies will provide



# EXPOSURE FROM AN IGNEOUS PHOSPHATE MINE OPERATION

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The International Atomic Energy Agency Safety Series 115 and European Union Council Directive 96/29/EURATOM have changed the profile of radiation protection completely, increasing the regulatory awareness of natural radiation and the industries involved. Three major industries have been identified in terms of their scope and the materials handled as industries requiring further attention. They are the Oil & Gas Industry, the Zircon Industry and the Phosphate & Fertilizer Industry. The Phosphate and associated Fertilizer industry has an added complication, because it has two distinct sources of raw material, e.g. being of igneous or sedimentary origin. The sedimentary material has a dominant  $^{238}\text{U}$  series, with activities ranging from 1 Bq/g per isotope to as high as 5.7 Bq/g per isotope, but with negligible  $^{232}\text{Th}$  content. The igneous material of the Phalaborwa Complex have fairly low levels of  $^{238}\text{U}$  and its associated daughters, (less than 0.15 Bq/g per isotope), but with elevated levels of  $^{232}\text{Th}$  when compared with the sedimentary material. This paper will focus on the mining operations of an igneous source located in South Africa.

The mine involved received a nuclear authorisation in 1993 under the auspices of the Nuclear Energy Act, No 131 of 1993 and in the following years completed both occupational and public risk assessments as required by the authorisation. This paper place emphasis on the public risk assessment completed in 1999, the re-



sults of the subsequent routine monitoring program and expand on some of the practical problems the company had to deal with.

The public risk assessment was conducted in an integrated manner, assessing doses to members of the public via the atmospheric, aquatic and secondary pathways by the Nuclear Energy Corporation of South Africa and the subsequent routine monitoring program results evaluated and reported to the regulator by the company itself.

In conclusion it will give a brief description of the current monitoring program with a mention of possible future projects.





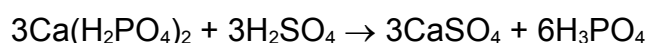
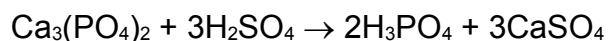
# RADIOLOGICAL RISK ASSESSMENT OF PHOSPHOGYPSUM PLASTERBOARDS IN HOMES

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Naturally occurring radioactive materials, or NORM, became the focus of regulatory interest with the publication of International Atomic Energy Agency SS115 and the subsequent publishing of European Council Directive 96/29/EURATOM. As a result, strong focus was placed on the Phosphate and Fertiliser Industry not only in Europe, but also South Africa. Foskor, together with the Fertiliser Society of South Africa (FSSA) embarked on a national industry assessment program to identify areas of risk as to ensure an optimised and cost-effective management program.

Phosphogypsum is generated during the production process of phosphoric acid from phosphate rock. Simplified reaction equations are as follow:



The source material contains isotopes from the natural uranium and thorium decay chains that may or may not follow the gypsum in the process. It is reported



that approximately 80% of the Radium-226 follow the gypsum, whereas 86% of the uranium and 70% of the thorium are found in the phosphoric acid. It is common practice to use phosphogypsum in the building process, such as in the manufacture of plasterboards for houses etc. (Ceilings are typically made from plasterboard).

The following assessment utilises actual measurements and radon calculations to assess a first order exposure risk for members of the public, when residing in a dwelling with phosphogypsum plasterboard as part of the structure as well as to the responsible builder in terms of occupational exposure. The assessment identifies areas that may require further or more detailed investigation, depending on the level of detail required by the regulatory authority and compare it with a similar study conducted in Australia on sedimentary material.



# RAIL TRANSPORT OF IGNEOUS PHOSPHATE ROCK

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The third Research Co-ordination Meeting on the Development of a Radiological Basis for the Transport Safety Requirements for Low Specific Activity Materials and Surface Contaminated Objects (LSA/SCO) was held in Cape Town, South Africa on the 19<sup>th</sup> to 23<sup>rd</sup> of February 2001. It has been attended by the Chief Scientific Investigators (CSI) from Brazil, Canada, France, Germany, South Africa and the United Kingdom, with the IAEA acting as Scientific Secretary. In addition to the CSI's, Observers of South Africa and Japan participated.

A Phosphate Rock and Fertiliser Producer (South Africa) participated under observer status during the event and agreed to repeat the zircon sand study for igneous phosphate rock and -phosphoric acid to support the theoretical models although the material is strictly speaking not subject to the requirements of "ST1: Regulations for the safe transport of radioactive materials".

This paper represents a partial fulfilment of the commitment, reporting on (a) the occupational exposure of loading of the phosphate rock on the rail cars, transport and unloading in the harbour (b) public exposure during rail transport and interim storage in silos and (c) give some reference to undesired events, such as spillage. This risk assessment is based on actual measurements where possible and will only refer to modelling or theoretical calculations where no results are available.



# NORM IN BUILDING MATERIALS

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Specific activity ( $C_{ef}$ ) of building materials depends on amount of radionuclides in their composition. According to standards of radiation safety  $C_{ef}$  should not exceed a dose Bq/kg ( $C_{ef} = C_{Ra} + 1.31C_{Th} + 0.085C_K$ ). Gamma-spectrometric investigations in Azerbaijan demonstrated that  $C_{ef}$  for building materials manufactured from natural raw materials varied from 30 to 190 Bq/kg. Average  $C_{ef}$  for Azerbaijan is 69 Bq/kg.

Products made from artificial materials should not contain radionuclides. Investigations demonstrated that sometimes  $C_{ef}$  10 times exceeds standards and is 3800 Bq/kg at account of increased amount of radium. This strengthens hazard of their utilization. Cases are known when sources of ionizing radiation have been lost while manufacturing building blocks of cement and concrete. Being installed in dwellings they create hazardous situation. Utilization of high-radioactive industrial wastes to manufacture bricks or cement to lay floors was found out.

These data demonstrate urgency of constant control for quality of the products used in construction of buildings and dwellings.



The most effective and economic way of quality control of the building materials is gamma-spectrometric analysis. This method is scientifically grounded and passed through great practical expertise.



## NORM IN COAL

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While combusting coal outburst occurs into the atmosphere: a great amount of aerosol particles containing natural radionuclides (uranium, radium, thorium and products of their decay). Concentration of radionuclides in different coals varies in Bq/kg from 3 to 520 Bq/kg for  $^{238}\text{U}$  and from 3 to 320 Bq/kg for  $^{232}\text{Th}$ . Degree of radionuclide contamination of the environment depends on the original matter. For this reason it is necessary to study radioactivity of coals and to select that which contains the lowest amount of harmful matters. Concentration of radionuclides in cinder grows 3–4 times as compared with the original matter as while combusting coal the organic component burns out and concentration of radionuclides in cinder and slog becomes higher.

Thus, during a slight growth of amount of uranium in coal – 90.7 Bq/kg and in ash it might be 270.3 Bq/kg. Normally amount of uranium in ash varies 500–600 Bq/kg but sometimes it is 1742 Bq/kg and even 3055 Bq/kg. This results in a heavy contamination of the environment and causes problems with the removal of such ash.



Knowing amount of radionuclides in coal and in ash, one can calculate safety level of the environmental contamination and to increase efficiency of purifying to substitute radioactive coal by coal with lower amount of uranium.



# **RADIONUCLIDE CONTAMINATION OF THE NATURAL ENVIRONMENT OF ABSHERON PENINSULA (AZERBAIJAN)**

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The territory of Azerbaijan due to its geographical situation, the total combination of nature-climate zones and the presentation of industrial potential is the unique object of ecological researches.

One of the most ecologically problematic regions of Azerbaijan is Absheron Peninsula. The main factors of pollution in development of oil deposits are the oil and oil products. Normal radioactive background in the Absheron peninsula varies from 4 to 10  $\mu\text{R/h}$ .

The problem of pollution of oil-producing areas by the radionuclides of the natural origin arises recently. Polluted by radionuclides surfaces and oil industrial equipment are discovered at almost all exploration fields surrounding the large industrial centre – Baku city. There were determined anomalous areas where intensity of gamma-radiation reaches 600  $\mu\text{R/h}$ . Besides, within the Absheron peninsula there are iodine plants, the territory of which are also polluted by radionuclides of the natural origin.





The identification of the radionuclides showed that they were presented by the group of uranium-radium, the content of which reaches the high concentrations. One of the methods of reducing the ecological intensity on the oil fields can be the utilization of underground salt brines and, probably, their usage for the medical goals.

During the process of oil-gas fields exploration (especially in the offshore area of the Caspian Sea) it is necessary to take environmental protection measures before geologic-exploration works linked with the study of radiation background of the coastal-shelf zones and radioactivity of bottom sediments. At present there is a large experience of radioecological works that can be used in creation of radiation monitoring in Azerbaijan.



# MITIGATION METHODS IN SELECTION PLACES OF CONSTRUCTION SITES

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One of the main tasks in selection of construction sites is study of geologic structure. Tectonic faults within construction sites represent additional hazard for buildings. Radiometric methods allow to determine activity of fault dislocations. In radioactive fields of the earth crust only active tectonic dislocations and geodynamic zones manifest themselves.

While planning construction works it is necessary to locate buildings outside the hazardous areas. Results of surveys allowed to identify areas with increased radon exhalation. It mitigates risk of hazardous radiation of people. According to level of gamma-radiation areas of grounds of different lithologic composition are identified.

In developed countries (USA, Canada) radon investigations were conducted to evaluate high, middle and low level of radon exhalation. In these countries it is used to choose less hazardous zones. In Russia radon survey for engineer-technical construction in different areas has been conducted more and more intensive recently. This is also used for the studies of interrelation of distribution of ground radon and amount of radon in buildings.



These problems have been discussed in China recently with account of mitigation of radiation risk for population. This problem should be discussed in newly established Radon Centre (Poland).



# NATURAL RADIONUCLIDES IN RARE EARTH INDUSTRY AND THEIR ENVIRONMENTAL EFFECTS

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The environmental problem of naturally occurring radioactive material (NORM) in phosphate, petroleum and natural gas, and coal industries have been well known and widely investigated. But a few investigations were done for the occurring, migration and radiation problem to workers of NORM in rare earth industry. Uranium and thorium are associated with rare earth ore. As high as 7% and 0.25% of Th and U in rare earth ore was reported, all radionuclides in the uranium and thorium decay series are therefore exist in the rare earth ore, and distribute in the working place and waste in the rare earth industry. China has more than 70% of inventory of rare earth ore, and produced 60.000 tons of rare earth products every year, tens times of waste containing NORMs was therefore produced. It induced a relative environmental problem. The current status of NORM studies in rare earth industry in China is reviewed in this paper. The type and distribution of rare earth ore in China and their radiation level is first introduced. The radiation level of working place in some rare earth factories is also presented, finally the concentrations



of U, Th and their long-lived daughter radionuclides in the waste and their environmental effects are reviewed.



# CRYING WOLF! HOW TO DEAL WITH PRESUMED RADIOACTIVE CONTAMINATION

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Reports concerning the occurrence of malignancies and cancer cases among the population caused by the presence of radioactive materials in the environment are not unusual in Brazil. The country's Nuclear Energy Commission, CNEN, is always assigned to launch an investigation in such cases. As a result, human and financial resources are usually committed in operations that, more often than not, end up by discovering that claims are not related to radiation. This time, alarmist reports from the small town of Lajes Pintadas, in the backlands of the Brazilian north-eastern state of Rio Grande do Norte, concerning the occurrence of stillborn and malformed children and of cancer cases among the adult population, reached the state capital. A team from the Regional Nuclear Sciences Centre, CNEN's institute for the north-east, was dispatched to Lajes Pintadas to look for the effective presence of environmental radionuclides and to investigate the reported occurrence of malignancies caused by radiation. Due to the well-known dangers related to launching an investigation on a subject so controversial and prone to misinterpretation by the media and the public, a novel approach, both in technical and philosophical terms, was attempted and that proved to be crucial for the success



of the whole undertaking: two years on, no more reports on health problems in Lajes Pintadas were published.



# ENVIRONMENTAL RADIOLOGICAL IMPACT BY A FERTILIZER COMPLEX IN THE EBRO RIVER (SPAIN)

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In this work we report the distribution of U and Th series radionuclides in the disposal area located in the vicinity of a fertilizer production complex in the Ebro river. This industry produces dicalcium phosphate (DCP), used as a source of calcium and phosphorus for domestic animals. The wastes are released into 5 dumping areas in the Flix hydro electrical reservoir, producing the presence of 5 sedimentary lobules near the river shore. A sediment core (longitude circa 1.5 m) was collected from each lobule. The concentrations of natural radionuclides determined by  $\gamma$ -spectrometry, clearly showed the enhancement of concentrations in the sediments since the beginning of production in 1973. The activities observed ranged from  $^{238}\text{U}$  (42–11 000 Bq kg<sup>-1</sup>),  $^{210}\text{Pb}$  (32–5600 Bq kg<sup>-1</sup>),  $^{226}\text{Ra}$  (21–9900 Bq kg<sup>-1</sup>),  $^{232}\text{Th}$  (10–163 Bq kg<sup>-1</sup>). These concentrations may be of concern as it is well known that radionuclides such as  $^{210}\text{Pb}$  can be accumulated by certain aquatic or-





ganisms. However, concentrations in water samples collected downstream, until the Ebro delta, have not shown the presence of enhanced levels of  $^{226}\text{Ra}$ .



# ASSESSMENT OF NORM IN SAUDI ARAMCO FACILITIES

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It has been established that Naturally Occurring Radioactive Materials (NORM) accumulates at various locations along the oil/gas production process. Components such as wellheads, separation vessels, pumps, and other processing equipment can become NORM contaminated. This can create a potential radiation hazard to workers and the environment.

Saudi Aramco Environmental Protection Department initiated a program to identify the extent, form and level of NORM contamination associated with their operations. This paper provides an overview of Saudi Aramco's NORM assessment program and highlights lessons learned and knowledge gained.



# THE $^{238}\text{U}$ – $^{234}\text{Th}$ EQUILIBRIUM IN SOIL SAMPLES

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At gamma-spectrometric measurements on environmental samples the  $^{238}\text{U}$  activity is determined from the gamma-lines of its first daughters  $^{234}\text{Th}$  and  $^{234}\text{Pa}$ . The 24.1 d half-life of  $^{234}\text{Th}$  might be in principle sufficient to disturb the  $^{238}\text{U}$ – $^{234}\text{Th}$  equilibrium in environmental samples. In few recent articles the possibility of quick  $^{238}\text{U}$  determination by means of gamma-spectroscopy was challenged on this base. In the paper we present the results of gamma-spectroscopic measurements on soil samples performed immediately after sampling and about 1 year after the sampling. The  $^{238}\text{U}$ – $^{234}\text{Th}$  equilibrium in fresh samples is discussed and the limits of the accuracy of quick  $^{238}\text{U}$  determination are presented.



# RADIOACTIVITY OF THE FOOD AT NOVI SAD MARKETS

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Foods bought at Novi Sad markets were investigated by gamma-spectrometric measurements. Milk and dairy products, cereals and cereals products, fruit, vegetable, meat and meat products, tea, herb, salt, mineral water, cacao etc. were collected. The activity concentration of 20 radionuclides in each sample was measured for 50 000 s. The average radiation ingestion dose is estimated only for radionuclide  $^{40}\text{K}$  for all foodstuff groups and the results are compared with the results of earlier measurements.



# NATURAL RESTORATION OF A SPANISH ESTUARY HISTORICALLY AFFECTED BY ANTHROPOGENIC INPUTS OF NATURAL RADIONUCLIDES

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The Odiel estuary, placed in the southwest of Spain, has been historically affected by the waste releases of several factories devoted to the production of phosphoric acid and phosphate fertilizers. A portion of the wastes was directly released into the Odiel River and other portion, about 80%, were stored in air opened piles, which could be affected by tidal processes. It is well known that these wastes are clearly enriched in radionuclides belonging to the uranium-series; being quite well documented the clear radioactive impact produced by them in several compartments of this estuary.



But the waste policy of the commented factories drastically changed in 1998. Since this year all the wastes are stored in well-protected piles without interaction with the surrounding environment. For instance, the water used for transferring these wastes is recycled. The consequences of the new wastes treatment relatives to the radioactive evolution of the estuarine environment are the main objective of this work. For that, we have studied the time evolution of activity concentration of several natural radionuclides in the estuary.

According to the previous studies, which have described the radioactive impact before 1998, the natural radionuclides more relevant are  $^{226}\text{Ra}$ ,  $^{238}\text{U}$ ,  $^{234}\text{U}$  and  $^{210}\text{Po}$ . In order to analyse, since the interruption of the releases until today, the time evolution of the activity concentrations of such radionuclides in the estuary, water and sediment samples were collected in three different campaigns (1999, 2001 and 2002). These results as well as their activity ratios are shown in this work and compared to the observed ones before 1998, when the releases occurred, showing that the contamination by natural radionuclides of the estuary is clearly decreasing nowadays. According to this tendency, the activity concentration enhancements caused by phosphoric factories, and consequently their radioactive impact, can be neglected in few years.



# TENORM REGULATORY FRAMEWORK IN BRAZIL

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Mining activities in Brazil are spread all over the territory. Many of these industries are suspected to cause undue exposures of workers and members of the public. Because of that, the Brazilian Nuclear Energy Commission (CNEN) through the Institute of Radiation Protection and Dosimetry (IRD) carried out a comprehensive investigation work to detect the most important ones, in respect to potential radiological impacts, and formulate the basis of a national regulatory framework to deal with the problem.

The work selected some ore processing industries, e.g., coal, niobium, phosphate and gold mining industries to be investigated. In parallel, the situation of the gas and oil industries, as well as from other industries like mineral sands and tin processing plants have been investigated by means of non-regulatory inspections.

Work methodology consisted of the examination of the individual operational process. Residues and liquid effluents were sampled from every single release point. The working places were also characterized by means of dust sampling, radon and gamma radiation measurements. The mobility of radionuclides from the wastes was assessed as well as the overall exposure of members of the general public, both in present (operational) and future (post-operational) scenarios.



The collected data permitted the construction of a robust data-base indicating the occurrence of major areas to be addressed in terms of remediation/clean up activities.

The results allowed the regulatory authority to issue a Regulatory Standard with the Brazilian Department of Mineral Production (DNPM) – CNEN.NE.4.10 ([www.cnem.br](http://www.cnem.br)). The standard divides the installations in three categories according to the total activity concentrations in the ore (or any residue) and the radiological dose. The first category encompasses industries that show activity concentrations above  $500 \text{ Bq.g}^{-1}$  and doses above  $1.0 \text{ mSv.y}^{-1}$ ; the second encompasses installations that lies down in the range between  $500\text{--}10 \text{ Bq.g}^{-1}$ , and the third one involves industries where the concentrations are smaller than  $10 \text{ Bq.g}^{-1}$  and doses bellow  $1.0 \text{ mSv}^{-1}$ . Different requirements are made to industries lying down in these ranges. As a result, it is expected that many of them will have to adopt remediation/clean up strategies that will require the assistance of specialized technical work.





## THE FIRST RADON MAP OF VOJVODINA

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Radon is a naturally occurring radioactive gas. Radon is the alpha emitter and decays to short-lived daughters ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ ). Radon partially decays in material where it has been generated and partially moves rapidly by concentration-driven diffusion into the open air. Underlying soil is the main source of indoor radon. However, building's materials also may contribute to elevated indoor radon concentration, especially in combination with low ventilation. In this paper the results of the first radon mapping in Vojvodina are presented. Indoor radon activity concentration in air has been measured at the whole area of the province Vojvodina (on about 2000 locations) by plastic track detectors CR-39. On the base of the obtained results, the average indoor activity concentrations of  $^{222}\text{Rn}$  for individual municipalities and for the whole province of Vojvodina ( $144 \pm 120$ ) Bq/m<sup>3</sup> were estimated.

Almost 20% of the measurements are over the 200 Bq/m<sup>3</sup> and 4% of the measurements are significantly elevated indoor radon concentrations. The results are above the expected values for the plains districts, despite the normal concentra-



tions of  $^{238}\text{U}$ , maybe due to underground waters. The dependence of indoor radon on the location and type of the houses and flats is discussed.



# EVALUATION OF OCCUPATIONAL RADIOLOGICAL EXPOSURES ASSOCIATED WITH FLY ASHES FROM FRENCH COAL POWER PLANTS

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The French Ministry of Health is preparing a series of decrees for transposing into French regulation the Euratom 96/29 Directive title VII concerning in particular the work activities where natural radioactive substances are handled and used, but not for their radioactive, fertile or fissile properties. Coal power stations belong to a preliminary list of industrial sectors potentially concerned by this transposition and the decrees' calendar of preparation includes a consultation period allowing to collect information about the possible dosimetric impacts on various population groups of these industrial sectors. At the request of the two French operators of coal power stations, Electricité de France (EDF) and the National Company of Electricity and Thermics (SNET), CEPN has evaluated occupational radiological exposures resulting from the industrial activities which bring into play fly ashes produced by the French coal power stations.



In a first step, the various stages of the French fly ashes cycle were studied, namely their production, handling and transport, storage, and recycling (mainly in building materials and road constructions). In a second step, reference groups of workers likely to receive significant doses were identified. Finally, a series of exposure scenarios, aiming to be both conservative and realistic, were described on the basis of realistic exposure data (when available) together with generic values and simplifying assumptions. In the absence of dosimetric measurements, individual exposures were evaluated using appropriate models for external irradiation, dust and radon inhalation, ingestion and transfer in the biosphere.

Estimated annual individual doses range from fractions to hundreds of microsieverts per year and maximum doses are associated with situations where external contribution is largely dominating (tailings works, road construction). Sensitivity analyses were performed to evaluate the impact on dose estimates of possible variations of the calculation parameters. This study should provide both industrial and regulatory bodies with a methodological approach enabling to pinpoint situations in the French context that may be calling for a particular attention in terms of radiation protection.

This paper summarises the methodology of the assessment as well as the characteristics and individual doses results associated with the most important scenarios.



# RECYCLING OF $^{232}\text{Th}$ CONTAMINATED TUNGSTEN SCRAP

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In Krefeld, Germany, Siempelkamp operates a melting plant to treat steel scrap contaminated with natural radioactivity, mercury and other chemical and toxic substances. The so-called GERTA plant is licensed according to the German BIm-SchG (federal law on protection against environmental pollution).

The facility is equipped with a net frequency induction furnace and has an annual capacity of 2000 t/a. The purpose is to produce a final metal product, which is completely free of all contamination and can be released to the steel cycle without any limitations.

So far, the input has been carbon or stainless steels with contamination of  $^{226}\text{Ra}$  as main nuclide in case of NORM contamination or elementary mercury from oil and gas producing facilities or from chlorine electrolysis.

In a recent project, more than 130 t of tungsten and molybdenum contaminated with radioactive  $^{232}\text{Th}$  from the production of welding electrodes, have been successfully decontaminated and have been converted into a Fe-W-Mo-alloy, which could be sold as a high value product. By this way, up to 25 wt-% of



tungsten/molybdenum could be dissolved in an iron melt and the thorium-oxide could be transferred to the slag.

The slag can be released for recycling, if the 1 mSv/a-criterion for workers and population according to the current German radiation protection ordinance is fulfilled.

In an expert opinion it was ascertained, that according to the typical distribution of throughput in the GERTA plant, specific activities up to 65 Bq/g  $^{232}\text{Th}$  and 87 Bq/g  $^{226}\text{Ra}$  can be accepted in the slag.

This paper describes the “tungsten project” in terms of metallurgy, radiation protection and legislation.



# ON THE MEASUREMENT OF $^{40}\text{K}$ IN NATURAL AND SYNTHETIC MATERIALS BY THE METHOD OF HIGH-RESOLUTION GAMMA- RAY SPECTROMETRY

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In the measurement of Natural Occuring Radioactive Material (NORM) the concentrations of  $^{40}\text{K}$ ,  $^{232}\text{Th}$  and  $^{238}\text{U}$  are measured by passive gamma ray spectrometry with either HPGe or NaI(Tl) detectors.



$^{40}\text{K}$  is measured through its 1460.8  $\gamma$  line. However till now it was ignored that this line is mixed with the 1459.2 keV line of  $^{228}\text{Ac}$  from the the chain of  $^{232}\text{Th}$ . A correction must be made for this contribution.

All the data in literature has wrong data about the concentration of  $^{40}\text{K}$  although in many cases the error is less than 1%. However there are cases of considerably higher errors. It should be emphasized that even if the correction in  $^{40}\text{K}$  concentration is large the correction of external dose index is negligible due to the higher weighing factor for  $^{232}\text{Th}$  than for  $^{40}\text{K}$  (by at least a factor of 10).





# SOIL CONTAMINATION IN A RURAL SITE USED FOR RARE EARTH INDUSTRY BY-PRODUCT DISPOSAL

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In the past the processing of monazite aiming production of rare earth elements was performed in São Paulo state. This industry produced large quantities of byproducts and wastes: cake II (uranium and thorium hydroxides) and mesothorium cake ( $Ba(Ra)SO_4$ ). At that time the National Commission of Nuclear Energy – CNEN didn't have regulations for this kind of activities. At the moment CNEN is preparing regulations regarding NORM processing industries and also for intervention situations.

Between 1975 and 1981, 3500 tons of cake II was stored in seven rectangular concrete pits three meters deep, surrounded by 30 cm concrete walls and floor, which were built for this purpose in a country area of São Paulo state, a region of farms. A river flows across the property into the public water supply of a city, 12



km far from the site. Activity concentrations of  $^{228}\text{Ra}$  varying from 0.5 to 50 Bq/g were identified in the soil by a preliminary survey, which indicates that the site was contaminated by radionuclides of the  $^{232}\text{Th}$  series from the by-products and wastes. The inhabitants of the region are very concerned about the situation.

The purpose of this paper is to identify and characterize the extension of the contamination in the soil and to derive guideline concentrations with the goal of the site remediation. Besides, an assessment of public risk perception has been performed. This assessment will give support to introduce the public participation in remediation process and establish a work methodology, in order to obtain a more effective communication between regulator and society.



# PASSIVE ANTINEUTRINO SOUNDING OF SPATIAL DISTRIBUTION OF RADIOACTIVE ELEMENTS IN THE EARTH DEPTHS

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The method is offered of the Earth depth antineutrino sounding with the help of antineutrino registration by several detectors located at different points of the Earth surface.

The modern knowledge about structure, element composition, physical condition of plutonic layers of the earth crust, the mantle and core of the Earth are insufficient for the solution of many problems. For example, the information about the element structure of the Earth mantle and core can help during refinement of an earth type planet formation models its evolution. At the same time, the knowledge of quantity and spatial distribution of radioactive elements in the Earth depth will enable to update a heat balance of the Earth, that can essentially influence on the development of climate models and methods of forecasting of climate change, and also, in general, on our ideas about a nature of energy-mass processes in plutonic entrails of the Earth. The especially practical problem of obtaining of more detail contours of radioactive elements fields is also important. All these problems are connected with determination of spatial distribution and quantity of radioactive



elements in the Earth depth, and its can be solved with the help of antineutrino sounding of our planet.

Except the type and quantity of nucleous, which emit antineutrino, the determination of a spatial distribution of these nucleous is very important. It can be made with the help of a passive detection and ranging methods, which have reached high perfection during the last years due to practical requirements of medicine, military applications and development of computer equipment. The problem of antineutrino sounding of the Earth depth is similar with some differences to problems of a medical tomography and problem of a passive electromagnetic or sound detection and ranging of moving objects.

The technology of passive sounding requires large interdetection base. The size of this base should be as greatest as possible in order to achive the maximum accuracy of measurements. Therefore for sounding of the planet depth, it is necessary to place detectors at the opposite points of the globe, for example, one detector is placed in Antarctic Continent, second – in Europe, third – in Asia and so on. All detectors should be connected by the common electronic circuit, which will ensure correlation measurements.

The arrangement of one or several detectors in Antarctic Continent or Greenland can be useful from the several points of view. First, it can be placed under a thick enough layer of ice, which will ensure the space radiation detector screening. Secondly, ice can serve as the detector material, and the size of the detector in this case will limit only by available quantity of photomultipliers and detectors of neutrons, so the size of the detector will be determined by the accessible cost and experimental expediency.

Thus, the organization of antineutrino-correlation experiments with the purpose of passive sounding of the Earth, on the one hand, opens the exclusive capabilities of the direct identification of a radioactive element spatial distribution in the Earth mantle and core, and on the other hand, can help to solve some relevant problems during the refinement of the planet formation models. For example, to find out the connection between the field of magma speed and the complex rotation and transmittion motion of continental plates.



## STATUS OF RADON DOSIMETRY IN ZAMBIAN UNDERGROUND MINES

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The Zambian economy has significantly relied on copper and cobalt mining in the relatively uraniferous Katanga Basin with a potential hazard of radon for over 50 years now. The radon preliminary survey of Hayumbu et al (1998) showed that at least 30% of the 42 randomly sampled sites in 8 underground mines had radon levels over 1000 Bq/m<sup>3</sup> (IAEA, 1996).

This paper outlines on-going radon dosimetry activities in Zambian underground mines and presents radon grab sampling and personal measurements at one of the mines identified to have high radon concentrations during the preliminary survey.

### References

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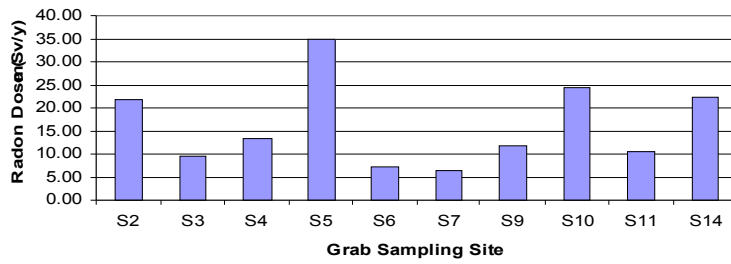


Figure 1. Radon Grab Sample Measurements of September 2002

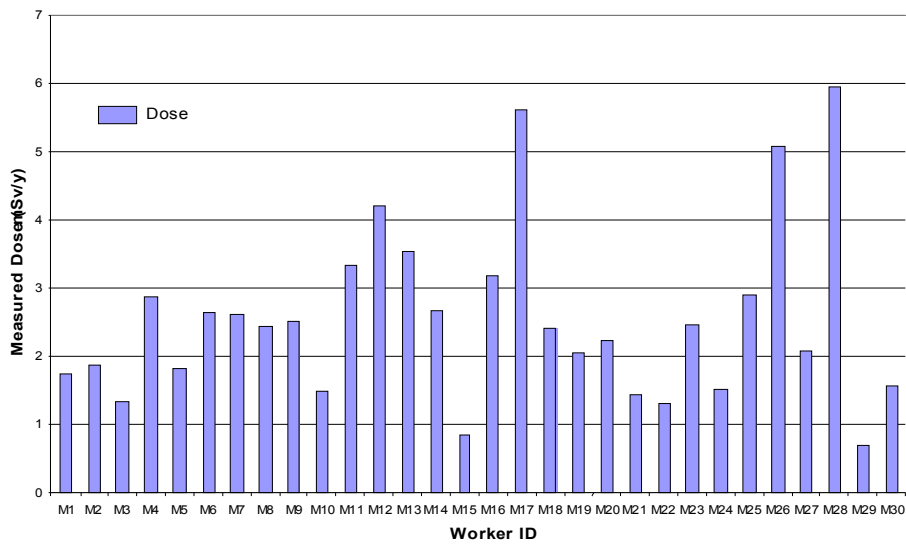


Figure 2. Radon Personal Measurements of September 2002 – June 2003



# THE RADON DOSEMETER COMBINING CHARCOAL CANISTERS AND TLD DETECTORS TYPE MCP-N (LiF: Mg, Cu, P)

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A method of measurement of radon concentration in air is based on high-sensitivity LiF: Mg, Cu, P (MCP-N, TLD Poland) thermoluminescent detectors installed in charcoal canisters [1].

To increase the radon measurement accuracy and to decrease detection threshold the new charcoal canister construction has been proposed. This makes possible to measure low radon concentration typically occurring in dwellings.

A calibration curve has been determined by exposing the canisters for 72 h in the calibration chamber IFJ-KR-600 with a radon concentration from 50 to 500 Bq/m<sup>3</sup>.

It has been found that in these conditions the signal registered by the TL detectors is proportional to the <sup>222</sup>Rn concentration and the lowest limit of detection is at a level of 50 Bq/m<sup>3</sup>.

The proposed method can be used in a routine measurements radon concentration in dwellings.



J. Bogacz, J. Mazur, J. Swakoń, M. Budzanowski, P. Olko MCP-N (LiF: Mg, Cu, P) TLDs for radon measurements with charcoal canisters. Rad. Prot. Dos. 101(1-4), 267-270 (2002).





# NORM AND DECOMMISSIONING OF NUCLEAR FACILITIES

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Recent experience in the United States has shown that NORM (naturally occurring radioactive material) can cause interference in the final status survey which is necessary to release a decommissioned nuclear facility from its license. Radiological assessments conducted in support of license termination often do not discriminate between nuclear materials which are part of the nuclear fuel cycle and those that are naturally occurring in the environment. Because of the greater linear energy transfer of alpha radiations, those naturally occurring isotopes that decay with the emission of an alpha particle are the cause of particular consternation. In addition, interpretation of data is confounded by the contributions of radiocaesium, iodine, and technecium contaminations deposited in the environment from nuclear weapons testing and accidents such as that which occurred at Chernobyl in 1986. Discussed are experiences and effective methods for discriminating between NORM and licensed materials during the application of field scanning and data reduction.



# IMPORTANCE OF SAMPLING IN RELATION OF THE GAMMA SPECTROSCOPIC ANALYSES OF NORM MATERIAL

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In the Netherlands problems arose with naturally occurring radioactive materials (NORM) and enhanced concentrations of NORM (TENORM; Technical Enhanced NORM) in the mid eighties of the last century and was due to discussions about reuse of fly ash in building and road construction materials.

Gamma spectroscopic analyses of NORM and TENORM samples were at that time still in its infancy. Nowadays different gamma spectroscopic analyse techniques exists all with their specific accuracy.

Nowadays state of the art techniques are able to assess the specific activity of uranium and thorium chains present in the variety of NORM and TENORM material with accuracies between 2 and 10%, even when daughters nuclides are not in equilibrium with their mothers.

This means in practice that the error due to sampling of NORM material has become and will be more and more important by the radiological characterization of samples of NORM and TENORM materials. This importance will also increase by implementing the EC directive L 159, 1996 in national legalisations of the Euro-



pean Community member states. The specific activity of samples has then to be calculated according to the weighted sum method as described in EC directive L 159.

An overview of the different (existing) gamma spectroscopic analyse techniques, from straight forward up to state of the art techniques, will be presented including their unique advantages and disadvantages.

Further special attention will be given to representative sampling of NORM and TENORM materials in practice, in order to minimize sampling error and thus minimizing the overall error. Applicable standards and guidelines will be discussed and also how to comply with these standards and guidelines in practice.



# FACTORS CONTROLLING MEASUREMENTS OF MASS RADON EXHALATION COEFFICIENT

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The determination of mass radon exhalation coefficient usually consists of measurements of radon contents in an emanation chamber containing a certain sample of investigated material as a function of radon growth time. The procedure seems to be simple, however results of radon exhalation coefficient measurement of the same investigated material from different laboratories usually differ. To answer the question, the authors attempted to investigate some factors such: sample mass, the ratio of a sample to emanation chamber volumes (volume ratio  $V_p/V_k$ ), sample granulation and moisture.

To investigate the above mentioned factors an emanation chamber of a volume of  $10 \text{ dm}^3$  and a Lucas cell with radiometer pylon AB-5 were used. The measurement procedure of the radon exhalation coefficient is described as follows: an investigated sample of a known mass and volume was closed tightly in the emanation chamber, then the growing of radon content in the chamber was measured. The radon contents were measured every 24 hours or so. The typical curve describing the relation between the radon content (N) in the emanation chamber and time (t)



is presented in Fig. 1. The curve of Fig. 1 can be presented by a well known equation

$$N(t) = a(1 - \exp(-bt))$$

where  $a = (E_m \cdot b)/m$ ,  $b$  is decay constants of radon,  $E_m$  is the mass radon exhalation coefficient [ $\text{Bq} \cdot \text{s}^{-1} \cdot \text{kg}^{-1}$ ] and  $a$  – sample mass [kg]. In Tables 1, 2 and 3, the measured values of radon exhalation coefficient are presented for samples of the different mass, different volume ratios  $V_p/V_k$ , different grain sizes and humidity.

The results given in Tables 1a & 1b show that the measured values of radon exhalation coefficient strongly depend on physical sample parameters. So they should be reported together with the measured values of mass radon exhalation coefficient. The trial of explanation of this phenomenon will be published by Chau et al (2004).

This work was partly sponsored by the Polish Radon Center

Chau N.D., Kalita K.J., Chruściel E., Prokólski Ł. 2004 „Factors controlling measurements of mass radon exhalation coefficient”, Journal of Environmental Radioactivity (under preparation).

Nezmal M., 2002, The international measurement of mass radon exhalation coefficient of some building materials. Unpublished report, Czech Republic, September 2002.

Table 1. Table 2. Table 3.

Mass [kg]	$V_p/V_k$	$E_m \pm 2\sigma$ ( $\mu\text{Bq} \cdot \text{s}^{-1} \cdot \text{kg}^{-1}$ )
0.05	0.0057	185±15
0.10	0.0140	189±12
0.20	0.0273	209±8
0.35	0.0412	215±7
0.50	0.0570	212±6
0.70	0.0800	175±8
1.00	0.1130	168±4

Grain sizes	$E_m \pm 2\sigma$ ( $\mu\text{Bq} \cdot \text{s}^{-1} \cdot \text{kg}^{-1}$ )
<1 mm	165±6
1–2 mm	167±5
2–4 mm	170±7
>4 mm	225±11
–	–
–	–
–	–

Humidity [%]	$E_m \pm 2\sigma$ ( $\text{Bq} \cdot \text{s}^{-1} \cdot \text{kg}^{-1}$ )
0	129±4
2	212±6
6	302±11
10	217±7
–	–
–	–
–	–

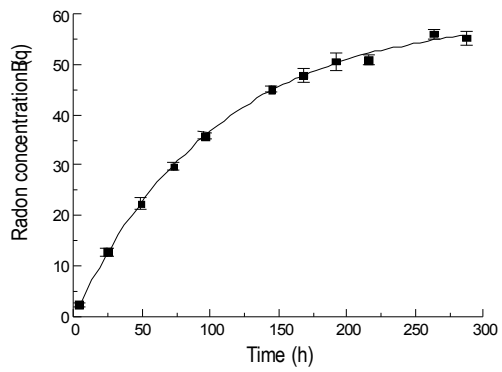


Fig. 1. The typical curve describing a relation between Rn content & time



# CONCENTRATIONS OF $^{222}\text{Rn}$ IN GROUNDWATERS FLOWING THROUGH DIFFERENT CRYSTALLINE ROCKS: AN EXAMPLE FROM ŚLĘŻA MASSIF (POLAND)

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Śląza Massif is situated in the south-western part of Poland, about 30 km SW of Wrocław, the capital city of Lower Silesia. The geological setting of the research area is typical of the Sudety Mountains. Different types of crystalline rocks, both of igneous and metamorphic origin, occur on the area of 25 km<sup>2</sup>. On the surface of this relatively small area, Lower Carboniferous to Lower Permian Strzegom̃ Sobótka Granite and Devonian Śląza Ophiolite are uncovered. The result is the occurrence of granites, gabbros, amphibolites and serpentinites, directly neighbouring on each other.



The author selected this area for determining the influence of rock type on the concentration of  $^{222}\text{Rn}$  dissolved in groundwaters flowing through crystalline rocks. The first stage of the research consisted of determining typical values of  $^{222}\text{Rn}$  concentration in groundwaters flowing through different types of rocks and describing the scale of seasonal changes in  $^{222}\text{Rn}$  concentration. In the next stage of the research, an attempt to apply  $^{222}\text{Rn}$  as one of the isotopic hydrogeochemical tracers of pathways flow of fissure groundwaters will be undertaken.

The results show that the highest values of  $^{222}\text{Rn}$  concentration (reaching 229 Bq/dm<sup>3</sup>) were observed in groundwaters flowing out of springs located within granite, whereas the lowest one (1.1 Bq/dm<sup>3</sup>) was noted in a spring located within serpentinite. The average  $^{222}\text{Rn}$  concentrations obtained in groundwaters flowing out of two springs within granite were 174 and 85 Bq/dm<sup>3</sup>, whereas the average values in two springs located within amphibolites reached 7.3 and 8.7 Bq/dm<sup>3</sup>. The average  $^{222}\text{Rn}$  concentrations in the springs flowing out of gabbro and serpentinite amounted to 7.4 Bq/dm<sup>3</sup> and 1.2 Bq/dm<sup>3</sup> respectively.  $^{222}\text{Rn}$  concentration in the groundwater flowing out of the spring located within serpentinites was stable during the whole year, likewise the discharge of the spring.  $^{222}\text{Rn}$  concentrations between  $1.1\pm 0.3$  and  $1.3\pm 0.3$  Bq/dm<sup>3</sup> were measured. On the other hand,  $^{222}\text{Rn}$  concentrations in waters flowing out of springs located within granite were characterized by distinct seasonal changes. The highest values (101 and 229 Bq/dm<sup>3</sup>) were noted in summer, when the discharges were low, whereas the lowest ones (70 and 127 Bq/dm<sup>3</sup>) were recorded in spring, when the discharges of both springs were high. In the remaining springs, radon concentration changes were of low scale and irregular character. The obtained results will be crosschecked with data obtained from other areas with similar geological structure, where crystalline rocks play the principal role.





# **RADON CHAMBERS – TECHNICAL DESIGN AND SOME CALIBRATION PROBLEMS**

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In the Natural Radioactivity Laboratory (at the H. Niewodniczański Institute of Nuclear Physics, PAS in Kraków) the special stand was designed and made for the calibration of radon detectors and for other research activities. The stand is equipped with two calibration chambers: IFJ-KR-200 (volume of 200 dm<sup>3</sup>) and IFJ-KR-600 (volume of 600 dm<sup>3</sup>), two certificated radon sources (<sup>226</sup>Ra activities: 21.6 and 52.3 kBq), a pump, a flow meter, a manometer and a control panel. The range of possible radon concentrations is from 45 Bq/m<sup>3</sup> to 244 kBq/m<sup>3</sup>. The computer software has been elaborated for planning and controlling the calibration procedure.

The active devices (AlphaGUARD by Genitron GmbH) are used for checking the obtained radon concentration inside the chamber. Two operation modes (diffusion or flow-through) were applied and compared. The results of testing the long-term tightness of the chambers are shown. The measurements of background radon concentration have been made and the methods of decreasing this level are also presented. The changeability of radon equilibrium factor F, as a function of radon



concentration and air humidity inside a chamber has been investigated and compared with the respective measurements performed in larger chambers situated in other laboratories in Poland.



# CALIBRATION OF SOIL PROBE FOR IN SITU PERMEABILITY MEASUREMENT

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Soil permeability is one of the important physical soil parameters for modeling radon transport from soil to indoor. Permeability relates flow ( $v$ ) through the soil pores to the pressure gradient ( $dp$ ). For in situ permeability measurement we applied a soil probe that was designed and made at the INP, PAS. Because permeability depends on velocity we calibrated our probe in laboratory conditions. For calibration we applied semi-empirical formula for permeability, depending on porosity, particle-size and soil water saturation fraction. We used two types of sand, because it is almost isotropic and homogenous medium.

As a result we obtained formula for shape coefficient  $W$  depending on  $v$  (velocity of fluid flow). The formula is a polynomial regression

$$W = A + B_1v + B_2v^2 + B_3v^3$$

where  $A$  and  $B$  are constants. For calculating permeability we use the following term

$$k = W [v / dp]$$





# INFLUENCE OF METEOROLOGICAL CONDITIONS ON RADON MEASUREMENTS IN SOIL – PRELIMINARY RESULTS

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Radon concentration in soil gas and its exhalation rate from the ground are very important factors that influence on indoor radon concentration and, in consequence, human health. Both these quantities depend not only on soil parameters (e.g. porosity, humidity, permeability, radium content, etc.) but on meteorological conditions as well. Since autumn 2003 the systematic measurements concerning radon exhalation rate from the ground and radon concentration in soil gas have been performed in the Natural Radioactivity Laboratory on a special “Radon Study Field” which is located on the Institute grounds. The continuous registration of meteorological parameters is also carried on the spot by means of the Weather Monitor II (DAVIS production). The measurements of soil temperature and pressure difference on 30, 50 and 100 cm depths were performed additionally and the results were used for checking the influence of those parameters on radon exhalation rate.



The passive method (CR-39 detectors) and the ionization chamber AlphaGUARD PQ 2000 PRO (Genitron GmbH) are used for the measurements of radon concentration in soil. The AlphaGUARD gauge together with the special accumulation container serve for the determination of radon exhalation rate from the ground.

The paper presents the preliminary results of the investigation of the influence of some meteorological parameters (air pressure and temperature, humidity, precipitation, wind speed and direction) on radon concentration in soil and its exhalation rate.



# METHODS FOR ASSESSMENT OF THE OCCUPATIONAL EXPOSURE AT WORKING PLACES OF DIFFERENT TENORM INDUSTRIAL BRANCHES

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Within the TENORMHARM project\* one task concerns an assessment of the radiation dose to workers in different TENORM industries which are of some relevance within the participating EU and candidate countries.

This assessment is based upon information and data collected at the beginning of the project and, if existing, on national regulations and compared to EU standards. The most relevant industries of each participating country where TENORM occur were reported and compared, from which each participant selected one branch where an occupational exposure of more than 1 mSv/yr could be expected. Subsequently, the calculation of the radiation dose was carried out independently by each participating country. Also the real situation at the considered working place, as respiratory protection or indoor air cleaning measures, was considered.

The results of the dose calculation obtained by each contractor for the selected exposure scenario was compared with the same or a similar scenario described in Radiation Protection 107. This comparison demonstrated, that the applicability of

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\* International group carrying out FP-S EURATOM Project „TENORMHARM”



reference levels for regulatory control of workplaces given in RP 107 was limited because the considered scenarios used, do not reflect sufficiently the recent work conditions. Moreover, any scenarios described by the contractors had no applicable counterpart in RP 107.

The results of this comparison will be presented and proposals for realistic dose assessments at different working places where TENORM occur will be given.

Acknowledgement:

The work is funded and carried out in the EU 5<sup>th</sup> FP, Nuclear Fission and Radiation Protection, Contract No FIGM-CT-2001-00174.





# DETECTION SYSTEM FOR SURVEY OF THE PROFESIONAL EXPOSED PERSONAL

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The individual control at the internal and external irradiation is necessary for the entire personal who work in the nuclear ranges and in all the domains where are used radiation sources.

In our country, the accredited work for the radiological survey at the external irradiation is for the dosimetric film. In the countries with evolved nuclear activities, the survey of personal is made with dosimetric film and thermoluminescent dosimeters for evaluation as precise as possible of the equivalent dose and her evidence.

The thermoluminescent multifunctional dosimeter with thermoluminescent detectors type TL-100, obtained in our department, respectively LiF: Mg, Ti is one of the passive system of detection which can be used for comparing the dosimetric film.

In this paper are presented experimental data compared between the two system of detection. The data are: the dosimetric answer for different values of dose and errors of determinations are in acceptable limits.



# **THE PROPOSAL OF THE SWEDISH COMMITTEE ON MANAGEMENT OF NON- NUCLEAR RADIOACTIVE WASTE (IKA) AND THE IMPLICATIONS FOR THE MANAGEMENT OF NORM AND STORAGE OF NORM WASTE**

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The Swedish Government Committee on Management of Non-Nuclear Radioactive Waste (IKA) has the commitment to propose regulations regarding NORM to be included in the Swedish Radiation Protection Law and the Radiation Protection Ordinance.

The production, handling and waste storage of such products has earlier not been subject to legal regulations, but is now to be included in the Radiation Protection Law and Ordinance.

Waste that will be subject to the new regulations are for example, ashes containing NORM and ashes of biofuel containing cesium-137 from the Chernobyl accident, uranium-rich skarn from mining of iron ores (even if the mine was abandoned many years ago), gypsum waste from phosphate production, alum shale burnt to extract oil (there exist more than 50 million m<sup>2</sup> of such material), filters and



sludge from water treatment equipment and pipes and pumps contaminated with radium due to precipitation from water.

The Committee proposes that the producer that utilizes raw material containing naturally occurring radioactive substances shall have the full responsibility for future depositing of the waste. The responsibility shall include a fee to a new Government fund to cover the total costs for management and future storage of the waste. This fee is to be paid by the producer of the waste material.

In case the waste consists of material from old use of natural radioactive material with no present owner, e.g. uranium use for colouring ceramics, the fund may in some cases cover the cost. This might also be the case when the disposal costs are unreasonable high for a private person, e.g. for disposal of resins used for treatment of water, which is contaminated with so high concentrations of uranium or radium that it has to be handled as radioactive waste. Radioactive waste from old abandoned mines, e.g. uranium mineralised waste rock at old iron mines or alum shale excavations where no present owner has the responsibility for the waste are proposed to be regarded as contaminated sites and thus entitled to the same Government funds that are used for restoring land contaminated by chemical substances.



# SEQUENTIAL EXTRACTION OF URANIUM ISOTOPES IN SEDIMENTS FROM AN ESTUARY HISTORICALLY AFFECTED BY ANTHROPOGENIC INPUTS OF NATURAL RADIONUCLIDES

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Riverbed sediments collected from an estuary historically affected by several anthropogenic activities, particularly by uranium-series radionuclides enriched wastes discharged by several phosphate fertiliser plants, have been operationally speciated using a Tessier-based selective-leaching procedure. <sup>234</sup>U and <sup>238</sup>U content in each selective fraction were determined by alpha-particle spectrometry after radiochemical isolation and electrodeposition onto stainless steel planchets.

These studies were performed in the uranium-polluted estuarine sediments one year after the phosphate fertiliser discharges stopped. The aim of this work was to obtain useful information about the operational forms in which this element re-



mains associated with the sediments at this time. Our results show a significant fraction of uranium radionuclides associated to operational forms that could be related to weak forms of adsorption to the sediments. Consequently, it is possible to foresee a potential remobilisation of uranium contamination from sediments to the aqueous phase in the future under normal environmental conditions.

Keywords: U-isotopes,  $\alpha$ -particle spectrometry, speciation, sediments, TENORM.



# ASSESSMENT OF THE RADON CONTRIBUTION FROM MINING SITES TO THE GEOGENIC ENVIRONMENT

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An issue of NORMs in mining areas is the mean 'Rn-exposure' assignable to surface mining residues. Outdoor Rn-decay product concentrations (at breathing height), compared with elevated indoor levels, are relatively low and they fluctuate widely with time and location. Differentiating a mining contribution of 'Rn-exposure' from a geogenic exposure is a challenging metrological task, particularly in the later stages of remediation. The interacting vertical and horizontal atmospheric air exchange and the Rn exhalation, largely varying with moisture content of the ground, cause widely varying Rn concentration patterns that are influenced by the weather on short and long timescales. The combined effect of varying source and of varying air exchange can be split into its components by time-resolved concentration measurement of the decay chain members of Rn, since at a given time the local ratios of concentration of the chain members are a function of the air exchange only. From the actual concentrations and the air exchange the source term can be calculated. The concentrations of the short-lived decay products of Rn can be measured continuously far more accurately per unit time than the Rn gas concentration. The Rn decay product ratios, and thus air exchange rates, can be assessed continuously with a time resolution of the order of 30 minutes or less.



Since the main exposure contribution derives from a few high, short-lived exposures, accurate time-resolved measurement of Rn decay product concentrations offers the optimal metrology for differentiating Rn-exposure contributions. True instrument calibration and minimal entrance losses in the decay product measurement are a requisite for attaining acceptable accuracy with an otherwise only roughly defined environmental source/sink compartment model.



# **NEW REGULATORY DEVELOPMENTS AND GUIDANCE IN THE EU WITH REGARD TO NORM**

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A framework for a regulatory system for the control of exposure of workers and members of the public to natural radiation sources was introduced in Council Directive 96/29/EURATOM 13 May 1996. The directive deals with basic safety standards for the protection of the health of workers and the general public against the dangers posed by ionising radiation. In particular Title VII of the directive deals with significant increases in exposure due to natural radiation sources. This applies to “work activities” or industries within which the presence of natural radiation sources leads to a significant increase in the exposure of workers or members of the public and which cannot be disregarded from the radiation protection point of view. In accordance with Articles 40 and 41 of the directive, each Member State is directed to ensure the identification of such work activities and to setting-up the appropriate means for monitoring exposure and applying radiation protection measures as set out in the directive.

To assist Member States with the implementation of the directive in general and Title VII in particular, the Commission has published a number of guidance documents. However, no guidance has yet been developed in relation to discharges





from NORM industries. In order to begin to address this, a study was carried out, the results of which have now been delivered. The aims of this study were: to review the regulatory framework within Member States regarding the implementation of Title VII of the directive with respect to effluent discharges; to provide guidance for a realistic dose assessment methodology for NORM discharges and to establish criteria allowing the rapid identification of effluent discharges potentially requiring regulatory control. The results of this study will be considered by the Group of Experts established under Article 31 of the Euratom Treaty with a view to advising the Commission on appropriate guidelines.

In addition, the Commission has awarded a contract with a view to developing a Network for stakeholders involved in NORM issues to share experience and to build a common understanding of the inherent problems and workable solutions. The Network will aim to identify and promote good practice, to enhance risk awareness and to create a common risk culture between stakeholders. It will act as an archive for relevant documentation that will be available to the members of the Network.



## **INDUSTRIES GIVING RISE TO NORM DISCHARGES IN THE EU – A REVIEW**

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The focus of this paper is the identification of industries giving rise to discharges or residues of naturally occurring radioactive material (NORM) that are potentially significant in terms of public exposure and therefore may require regulatory control as set out in Title VII of EU Council Directive 96/29/Euratom (Basic Safety Standards Directive). The industries have been identified and assessed as part of a study commissioned by the European Commission with a view to assisting Member States in implementing the aforementioned Directive in this area. Information was gathered largely on the basis of data supplied by regulatory authorities in Member States by means of a questionnaire as well as through published reports and direct contacts with organisations such as trade associations. It is evident that available information on wastes and discharges both in terms of volume and content are limited. Furthermore, there are few reliable monitoring data for radioactive substances probably as a consequence of the absence of previous regulatory control.



The industries studied in this paper include: fossil fuel power stations; oil and gas extraction; metal processing; phosphate processing and production; titanium oxide pigment production; rare earth processes and cement production. A brief description of the processes involved in each is presented including an analysis of the waste production pathways, activity concentrations, typical emissions and annual throughput, where available. It is noted that characteristics of discharges and residues, even from the same type of industry and production process, have been found to differ widely because of the variation in raw material used and processing methods. This is particularly pertinent in respect of differences in treatment of liquid wastes and off-gases prior to discharge.

It is possible that for some of the NORM industries studied, controls already in place to prevent non-radioactive pollution may have the additional benefit of controlling the radioactive discharges. However it must also be highlighted that restricting discharges may increase the amount of radioactivity retained in residues such as sludge and dust.



# THE RAPID IDENTIFICATION OF NORM DISCHARGES REQUIRING REGULATORY CONTROL – A POSSIBLE SCREENING METHODOLOGY

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The European Commission (EC) is mindful of the potential difficulties for national regulators in implementing a process of regulatory control as set out in Title VII of EU Council Directive 96/29/Euratom (Basic Safety Standards Directive) for NORM industries discharging into the environment. As a consequence, the EC commissioned a study to identify possible procedures for the rapid identification of such industries or work activities where members of the public are potentially exposed, with a view to producing guidelines. The results of the study are presented in this paper.

It is well understood that there is no simple relationship between discharge rate from

a NORM industry and dose to members of the public. However, with respect to



regulatory control, a detailed site-specific analysis may not be warranted when, on the basis of a conservative approach, it can be concluded that the discharges are of no radiological significance. This introduces the idea of NORM discharge screening levels which may be defined as estimates of the amount of activity discharged to the environment, which, if not exceeded, mean that it is very unlikely that members of the public would receive an effective dose above a defined dose criterion. NORM discharge-screening levels can be defined for each NORM release route and would be calculated using deliberately cautious assumptions. The derivation of such screening levels would comprise the following steps: definition of reference discharge situations; choices of models, exposure pathways and parameters; derived doses per unit discharge rate; dose criteria for screening levels and finally, discharge screening levels.

In this paper, discharge-screening levels are derived and presented. It is unlikely that below these levels a more detailed site-specific radiological assessment of the discharges would be warranted. It is recommended that a dose criterion be chosen for the screening level that is below the dose constraint for the facility. If discharge-screening levels are exceeded it is recommended that regulatory bodies verify the actual level of discharge, re-examine discharge conditions such as stack height or river flow, re-examine the existence of assumed exposure pathways and decide on the need for site-specific assessment.



# POLISH NATIONAL INTERCOMPARISONS OF MEASUREMENT METHODS OF $^{222}\text{Rn}$ CONCENTRATION IN WATERS

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The results of intercomparison measurements of  $^{222}\text{Rn}$  concentration in the water samples are presented. In the above mentioned measurements generally 11 laboratories from many Polish research and commercial institutions took part. Seven of them participated in 3 intercomparison measurements organized in the years 2001–2003. In the years 2001 and 2002 the intercomparison measurements were carried out in the Sudety Mountains (Szczawno Zdrój and Świeradów Zdrój health-resorts, respectively). During these two experiments water samples were taken from mineral springs, considered in Poland as medicinal ones. They are characterized by increased  $\text{CO}_2$  concentration (1.56–2.07 g/dm<sup>3</sup>), as well as different values of total dissolved solids (TDS between 0.18 and 2.97 g/dm<sup>3</sup>), radon concentration (between 9 and 985 Bq/dm<sup>3</sup>) and other distinct differences in chemical composition and physical properties.

In 2003 experiment measurements were carried out in water samples prepared in CLOR in Warsaw. The samples were produced by the aeration of a 10-dm<sup>3</sup> container full of tap water with air enriched by  $^{222}\text{Rn}$  coming from a PYLON source (502.5 kBq activity of  $^{226}\text{Ra}$ ). During all intercomparison experiments the concentration of  $^{222}\text{Rn}$  for 7 water samples (2, 3 and 2 in 2001, 2002 and 2003, respectively) was determined. Special care was taken to ensure the same conditions of collecting the water samples for each participant.

The laboratories participating in the experiments were applying different techniques to determine the concentration of  $^{222}\text{Rn}$  in the water samples: a liquid scintillation method, gamma detectors, Lucas cells and ionisation chambers.

The obtained results confirmed that keeping the containers with water samples closed tightly from the moment of collecting till the measurements was of a great importance.

It happened twice, in the different laboratories that a leakage in the measurement set-up (in the AlphaGUARD™ + AquaKIT™ system) resulted in the determination of lowered values of  $^{222}\text{Rn}$  concentrations. The best consistency of the results in all 3 intercomparison measurements was obtained by the laboratories using the



liquid scintillation method. It has been observed, that the number of laboratories that got consistent results (within error limits) is increasing from 55% in 2001 to 75% in 2003. This confirms the fact that the organised experiments meet the main task – improving the correctness of the results obtained in particular laboratories. Thus, it is necessary to organise such experiments in next years.

The intercomparisons in 2002 and 2003 were organized within the confines of the activity of the Radon Centre and with its financial support.





# APPLICATION OF INTERNATIONAL SAFETY STANDARDS TO WORK INVOLVING EXPOSURE TO NATURAL RADIATION

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One of the statutory functions of the International Atomic Energy Agency (IAEA) is to establish standards of safety for protection against ionizing radiation and to provide for the application of these standards to peaceful human activities. The International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (the BSS), published by the IAEA in 1996 and co-sponsored by several other major international organizations, contain the basic requirements to be fulfilled in all activities involving radiation exposure, including exposure to radiation of natural origin. In developing supporting standards and practical guidance on the application of the BSS, the IAEA has given progressively greater attention to exposure arising incidentally from work involving raw materials containing natural radionuclides. This paper describes the direction that this programme is taking in response to the needs expressed over the past few years by participants in various international meetings and conferences organized by the IAEA. A need has been identified for more data on activity levels, exposures and doses, as well as the factors influencing these parameters and governing the rela-



tionships between them in specific industrial processes involving naturally occurring radioactive material (NORM). Various data-gathering initiatives are described, including the gathering, interpretation and reporting of existing, reliable data for specific NORM industrial processes, and the formulation of a Co-ordinated Research Programme starting in 2004 to gather new data. A need has also been identified for practical guidance on the establishment of national systems of control over exposures to natural radiation, taking into account the very wide range of exposure scenarios and the necessity for pragmatism – that is, for radiation protection to be optimized to ensure that the degree of regulatory attention given to any particular situation is commensurate with what can be realistically achieved. The paper will describe the progress being made in this area in terms of conclusions to be drawn from existing guidance material, the programme for developing this guidance further in areas where uncertainties continue to be experienced, and the dissemination of this guidance in the form of training courses and other activities.



## A SPECIFIC STUDY CONCERNING NORM IN REFRACTORIES INDUSTRIES

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A specific study concerning refractory industries has been performed by a group of Italian Regional Environmental Protection Agencies together with the National Environmental Protection Agency. The aim of this study was to evaluate the environmental impact for some activities dealing with NORM.

In this frame, all steps of the standard working cycle are described: the beginning from the raw materials (zircon sands or semi-finished zircon silicates based components) and the end up to the finished products and the production of residues, dust and treatment water. A radiological survey has been carried out in two Northern Italy companies.



Activity concentrations measurements by gamma spectrometry (more relevant gamma nuclides of natural chains) and radiochemical analysis ( $^{210}\text{Pb}$ ,  $^{210}\text{Po}$ ) on raw materials, finished products, residues and treatment water were made.

Information and technical data related to the present working cycle have been collected by means of specific questionnaires, sent to most of the involved factories selected from the sector association's data base.

An estimate of the environmental pressure from the two companies and the whole sector itself is assessed by the combination of the radiological results of two companies survey with information from the questionnaires.



# GRAIN SIZE IN RADIOMETRIC MEASUREMENTS OF GROUND

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Known procedures of radiometric measurements of ground differ significantly in grain size of the sample. The range is 0.043–0.25 mm. To answer the question about the proper grain size of the sample to be measured one must address to the three types of radioactivity in grounds.

Natural radioactivity is distributed homogeneously through the bulk of sample. It is either nuclides in crystal structure of minerals (K in  $\text{KAlSi}_3\text{O}_8$ ) or isomorphic replacement in crystal structure (Ra in  $\text{BaSO}_4$ , Th in  $\text{ZrSiO}_4$ , U in  $\text{CePO}_4$ ). Model radiometric measurements of the same sample with grain size ranging from 0.125 mm to 2 mm do not show significant difference between specific  $\alpha$ - and  $\beta$ -activities in spite of increasing of specific surface. That is why there is no reason to grind monocrystal grains.

Artificial radioactivity is caused by atmospheric fallout presenting nuclides in dynamic form, or by “hot particles” which are commonly uranium and plutonium oxides (silicates) and also  $^{14}\text{C}$ . If we grind “hot particle” its fine subparticles will cover non-active grains of matrix that will lead to increasing of surface contamination and as a result – to increasing of specific radioactivity.



Secondary radioactivity is a result of changes of nuclides concentration in grounds owing to weathering, infiltration, precipitation and other natural processes. Both natural and artificial nuclides can be involved in these processes. Nuclides here occur preferentially in sorbed form on the grain surface as salts, hydroxides and complex compounds. This is another kind of inhomogeneity, not so sharp one. Grinding such grains increases specific surface, decreasing specific radioactivity. Thus the whole bulk of sample to be measured should be driven to monocrystal state, i. e. grain size does not have to be definite. In case if the sample of ground contains “hot particles” one must try to separate it to active and non-active parts as carefully as possible with two different subsequent measurements.



# TEST THRESHOLDS FOR ASSESSMENT OF POSSIBLE GROUNDWATER CONTAMINA- TION AT SITES CONTAMINATED WITH RA- DIOACTIVE MATERIALS

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At several sites in Land Brandenburg the ground is contaminated with natural radionuclides, especially thorium from former industrial processes. Such type of contamination is regulated neither in the Federal Ordinance on Soil Protection and Contaminated Sites (BBodSchV) nor in the German Radiation Protection Ordinance (StrlSchV). However, the European Basic Safety Standards Directive specifies that areas of former practice or work that are a source of permanent risk to people have to be demarcated and controlled. If necessary, intervention measures need to be carried out and access must be restricted. Therefore treatment of such



cases requires technical regulations and supporting instructions to facilitate correct decisions by the relevant authorities. Such regulations must conform with the existing regulations, especially the BBodSchV and guidelines of urban land use planning.

When assessing existing situations, the exposure path “groundwater” is of particular significance for any decision-making. In a study suitable criteria and instructions were derived in order to identify for this special path those areas that can be considered as uncontaminated and therefore exempt from further radiological investigations and from any restrictions concerning land use planning. The methodical approach used corresponds to the leachate forecast according to BBodSchV as applied for natural radionuclides.

Threshold values were derived from the relation between contamination of soil and resulting ingestion doses for children younger 1 year. The lower threshold “UWB” corresponds to a dose level of 0.1 mSv/a, the upper one “OWB” corresponds to 0.5 mSv/a. The following test thresholds “UBW” were obtained for a contamination close to or in contact with groundwater:

40 Bq/kg for the maximum activity of  $^{232}\text{Th}$  decay chain;

40 Bq/kg for maximum activity of  $^{238}\text{U}$  chain.

If the contamination is located significantly higher than the groundwater table (here: more than 1 m), then the values are:

1000 Bq/kg for the maximum activity of  $^{232}\text{Th}$  decay chain,

200 Bq/kg for maximum activity of  $^{238}\text{U}$  chain.





# NATURAL RADIOISOTOPE LEVEL DIFFERENTIATION IN ARABLE AND NONCULTIVATED SOILS AT ŁĘCZNA- WŁODAWA LAKE DISTRICT

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Pojezierze Łęczyńsko-Włodawskie district located on East of Poland is used as recreation ground but mainly it is an agriculture area producing ecological food. Due to lack of industry this area is relatively low contaminated. An exception is Bogdanka coal mine located 20 km north of Łęczna town at vicinity of Poleski National Park. Its activity, despite of the law and technical regulations observance may be disadvantageous for the environment in future. A potential hazard may give accumulation of natural radioactive elements in soil and bottom sediments, which results from increase of the storage field (up to 83 ha in future) and disposal of coal mine water to the Świnka river.

In the paper the comparison studies of the concentration of some natural radioactive isotopes in cultivated and unploughed area are presented. The samples were collected from 0–10 cm surface layer at seven points at Poleski National Park bor-



der. Concentration of gamma radioactive isotopes was measured by Silena-Canberra semiconductor (HPGe) spectrometer with Genie-2000 software for quantitative analysis of the nuclides. Obtained results enable estimation of health hazard by natural and anthropogenic  $^{137}\text{Cs}$  gamma radioactive isotopes.



# ATTEMPTS ON RADON EXHALATION RATE DETERMINATION FROM A WASTE-DUMP AT THE BOGDANKA COAL MINE USING THE PICORAD DETECTORS

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Bogdanka coal mine is located in Pojezierze Łęczyńsko-Włodawskie district at vicinity of Poleski National Park. From the beginning of the coal mine activity i.e. for 20 years waste rock is deposited close by. It mainly consists of clumps (about 88%) of a few centimeters grain-size, whereas sandstones, siderite and mudstones form the rest. Storage yard filled in about 30 percent covers 83 ha and is cultivated in part.

It is well known that deposited waste rocks of coal mines may be a source of radon.

The emission rate of this element from a spoil dump depends on many factors so direct measurements of an exhalation rate are the most reliable.



Passive detectors containing activated carbon, Picorad (Niton-Canberra-Packard), designed to the detection of indoor radon at stable concentration of this nuclide were used. An application of these detectors in open area may produce errors connected with higher humidity and varied concentration of exhaled radon.

The radioactivity of radon and its daughters was determined with liquid scintillation spectrometer by Quantulus (Wallac-Perkin-Elmer). Measurements were made in several points of the spoil dump including last deposited and cultivated sites. A calibration of the detectors with various concentrations of radon and humidity level was performed in a radon chamber in Central Mining Institute.



# DETERMINATION OF $^{210}\text{Po}$ AND $^{210}\text{Pb}$ FROM MARINE SEDIMENTS

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The sediments of the northeast Irish Sea are radioactively contaminated due to controlled discharges of low-level radioactive waste from nuclear facilities. Our core samples have already been well profiled with respect to U, Np, Am and Pu. The concentrations of these nuclides can be compared to Sellafield discharge declarations. In order to date the sediments, however, we must determine other radionuclides. The so-called  $^{210}\text{Pb}$  method is widely used to determine the accumulation rate of sediments in lakes, oceans and other waters. The age of sediment from a certain depth in a sediment core can be established from the accumulation rate. This paper describes the determination of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  with different methods. By experimentally comparing analytical methods we have chosen the one that is least time consuming and which has the best analytical figure of merit. Moreover, the core sediments have been dated and the results compared with the yearly discharge declaration of the facility.



# COSMOGENIC $^{22}\text{Na}$ AND $^7\text{Be}$ IN GROUND LEVEL AIR

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The cosmogenic radionuclides are produced in whole atmosphere, although the most intense processes goes in the stratosphere. One of such radionuclide is  $^{22}\text{Na}$ , which decays by beta plus decay with half-life-time of 2.6 year, end emits gamma radiation of 1275 keV. Another one is  $^7\text{Be}$ , which is also gamma-emitter (478 keV) and which half-life time is 54 days. Sooner or later all not gaseous matter attaches to the aerosols. From stratosphere cosmogenic radionuclides migrate to ground level air by means of different processes like diffusion, sedimentation, convection. Usually on ground level air the activity of  $^{22}\text{Na}$  is on the level of a fraction of single  $\mu\text{Bq}/\text{m}^3$  may be studied by filtration of high volumes of the air and application of low-level gamma spectrometry.  $^7\text{Be}$  is much more active, on the level of few  $\text{mBq}/\text{m}^3$ .

The Petryanov air filters from two ASS-500 aerosol sampling stations in weekly achieved sets exposed in Krakow during 2003 are analyzed for the presence of all gamma-emitters using a gamma-rays spectrometer with HPGe detector shielded with complex active and passive shields. Each sample contains aerosols from more than  $0.1 \text{ Mm}^3$  of air. In our previous study covering years 1996–2002 (in half



a year sets of samples) the cosmogenic  $^{22}\text{Na}$  shows a strong seasonal variation with significant different mean values activity concentration between  $0.333\pm 0.095 \mu\text{Bq m}^{-3}$  and  $0.137\pm 0.045 \mu\text{Bq m}^{-3}$ , for summer and winter, respectively [1]. Moreover, the activity ratio for two cosmogenic radionuclides:  $^{22}\text{Na}$  and  $^7\text{Be}$  showed also changes with statistically significant seasonal differences. The lower values were found during winters. The conclusion was that transport of  $^{22}\text{Na}$  from stratosphere to ground level air during summer seems to be so much effective, that result in kind of relative depletion of stratosphere of this nuclide. The aim of present investigation is to see the changes of  $^{22}\text{Na}$  activity and of  $^{22}\text{Na}$  to  $^7\text{Be}$  activity ratio in more detailed way within a year period.

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# APPLICATION OF LIQUID SCINTILLATION COUNTING TECHNIQUE TO GROSS ALPHA, GROSS BETA AND RADON MEASUREMENTS IN PORTUGUESE WATERS

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Natural water contains a number of alpha and beta emitters and the radiological examination such as the determination of the quality of drinking water requires measuring of mixtures of  $\alpha$  and  $\beta$  emitters. The recommendation of Portuguese law proposed limits values (0.1 Bq/l and 1 Bq/l) for gross alpha and gross beta activities, respectively. The main goal of this work was to develop, test and optimise new radiometric techniques for measuring mixtures of  $\alpha$  and  $\beta$  emitters in drinking waters. Liquid scintillation counting technique (LSC) has been optimized for the determination of these radiological parameters. We used a low background liquid scintillation system detector with the ability to discriminate between alpha and beta particles by pulse shape analysis (PSA). High counting efficiencies and lower limits detection were obtained. The first studies on the radioactivity content of some





bottled waters showed the limits values weren't exceeded. Water samples from taps, wells and springs have already been collected in old mining areas for further analysis. The results of gross alpha and beta measurements showed waters with more high activities.

This water samples collected in that areas are also enriched with dissolved Rn, produced in rocks containing uranium. Radon levels were determined by short lived daughter products using the LSC technique. The counter was calibrated with  $^{226}\text{Ra}$  standard solutions, measured one month after and corrected to correspond secular equilibrium of  $^{226}\text{Ra}$  with  $^{222}\text{Rn}$ . The counting efficiency was closer to theoretical maximum expected (500%). The LLD value for radon measurements was 0.195 Bq/l using the counting time of 60 min. Preliminary results will be described.



# EXAMINING THE NATURAL RADIOACTIVITY OF WATER SOURCES TO EVALUATE THE IMPACT ON SURROUNDING COMMUNITIES

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Natural occurring radioactive materials (NORM) are quite common in the South African mining and mineral processing industry, even so that uranium is produced as a by-product at certain gold and copper mining sites. Other major industries associated with NORMs are the coal, copper and heavy mineral sectors.

Legislation in South Africa addresses the allowed yearly radiation dose to registered radiation workers and the general public, and adheres to the most common internationally accepted standards of 20 mSv/a and 1 mSv/a respectively for these categories. The yearly dose is obviously the sum of the contributions from every individual nuclide from all possible sources. Accordingly, for members of the public, the individual sources are to be evaluated at the (South African) guideline level of 25  $\mu$ Sv/a. This imposes severe constraints on the radioanalytical laboratory to offer an affordable routine service due to the required sensitivity to analyse these NORMs and the variety of matrices involved.



Determination of all required parameters to evaluate the yearly dose is possible, although costly. Correlations of yearly dose with one or two chemical/radionuclide components will allow an affordable monitoring strategy. According to the evaluation of a survey performed in one of the gold-mining catchment areas in South Africa this seems indeed possible.



## THE UK

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As part of its periodic review of radiation doses to the population of the United Kingdom, the National Radiological Protection Board identified the need to estimate the radiological impact of the operation of non-nuclear industries which use or produce materials containing enhanced levels of naturally occurring radionuclides. A number of industries have been considered in the study. These include: coal-fired electricity generation; the steel production industry; the oil and gas industry; and rare earth industries. For each industry the radiological impact of the primary industry, the waste streams produced and, where applicable, the use of by-products were addressed. Doses to workers in the industries and members of the public were estimated as part of the study.



# **RADIONUCLIDE CONTAMINATION OF SURFACE WATERS, SEDIMENTS, AND SOIL CAUSED BY COAL MINING ACTIVITIES IN THE RUHR DISTRICT (GERMANY)**

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The discharge of highly mineralised mine waters with enhanced  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  activity concentrations has affected creeks, rivers, sediments, soils, and plants along the Lippe River and its tributaries.  $^{226}\text{Ra}$  activity concentrations were elevated in all water samples receiving mine water, with activity concentrations gradually decreasing with increased distance from the colliery due to dilution and chemical precipitation of radium with barium. Increased concentrations of radium and radium decay products were also measured in sediments and flood-affected soils. The sediments show an enrichment of  $^{226}\text{Ra}$  up to a factor of 750, while the contaminated soils “only” reach a factor of 10. In aquatic plants, a 4-fold increase in  $^{226}\text{Ra}$  activity concentrations was measured downstream of the discharge points. The contamination of the river banks and adjacent floodplain with radium is responsible for enhanced gamma dose rates, which, along with the incorporation of soil by playing children, provide potential radiation exposure to the public.



# PRESENCE OF NORM IN THE CZECH REPUBLIC

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NRI Rez participates in the TENORMHARM project within the 5th EU Framework Programme. NRI's effort is focused on evaluation of NORM inventory and assessment of radiation dose and health risks to workers and members of the public.

In case of phosphate fertilisers' production, three major factories ensure the Czech consumption at present. Nowadays, generally less radioactive raw materials are used such as yellow phosphorus, phosphoric acid, concentrate of Cola-apatite and AMOFOS. Only the concentrate of Cola-apatite contains higher quantities of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Th}$  (several tens of Bq/kg), but even in this case the resulted annual effective dose of workers does not exceed the regulatory limit.

Titanium dioxide pigments are produced only in one factory. Relatively radioactive ilmenite (approx. 140 Bq/kg of  $^{238}\text{U}$  and 80 Bq/kg of  $^{228}\text{Th}$ ) from Ukraine is used as the raw material. The final product (rutile- $\text{TiO}_2$ ) contains only negligible quantities of the mentioned nuclides. The majority of the radioactivity content thus remains in the various wastes – e.g. the content of  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$  reaches up to several 1000 Bq/kg in sludges.

Currently, one factory processes zirconium refractory material (EUCOR). Only zirconium-containing bricks (with specific activity approx. 1.700 Bq/kg of  $^{226}\text{Ra}$ ) dis-



carded from glass factories are used for production of EUCOR. The current consumption of approx. 200 tons of zirconium input material (bricks) means reprocessing of 340.000 kBq of  $^{226}\text{Ra}$ .

The coal exploitation and use represents one of the most important industries concerning NORM. The concentration of U in the Czech brown coal varies in the range 5–16.3 ppm and Th 0.2–30.2 ppm; the hard coal contains 2–9 ppm of U. The majority of the coal is used for the energy production. The U and Th concentrate mostly in the ash and slag (about 20 ppm of Th and <12 ppm of U); the concentration of all radioelements in gypsum produced in desulphurisation process is negligible.

The majority of oil and gas used in the Czech industries is imported from abroad. Measurements of samples of oil sludge from cleaning of IKL and Druzba pipelines and from of oil tanks exhibited only negligible concentrations (tenths of Bq/kg) of all radioelements. The similar range of activity has been found in samples from domestic oil exploitation.

Evaluation of radioactivity in sludges resulting from the 68 ground and surface water treatment installations was performed by NRPI. Average specific activity 200 Bq/kg of  $^{226}\text{Ra}$  was identified in ground water treatment sludges.



# THE PERIODICAL CHANGES THE CONCENTRATION OF INDOOR RADON

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Radon provides the largest contribution to the effective dose absorbed by the population inhabiting the region of normal radiation background. In Poland, the radon effective dose is about 39%. The radon concentration in the indoor air undergoes diurnal and seasonal changes. In order to analyze more accurately radon significance in the evaluation of radiological status of the population it is important to improve methods of the annual mean radon concentration evaluation, as the basis to assess the obtained effective dose.

One-year's radon concentration measurements were performed in 10, previously chosen, dwelling houses. Buildings qualified to the examination revealed the radon concentration of above 200 Bq m<sup>3</sup>. The integral method of Cr-39 trace detectors in diffusive chambers was used in the study. In each building, 3 detectors, changed every month, were exposed. All the buildings showed seasonal changeability in radon concentrations. The ratio of the radon concentrations during one-month's exposure to the mean annual concentration ranged from 0.16 to 2.35. Similar ratios were determined for 2-, 3-, and 6-months' exposure. The values of coefficients equaled from 0.2 to 2.3 for 2-months' exposure, 0.26 to 2.21 for 3-





months' exposure, and 0.36 to 1.54 for 6-months' exposure. Most buildings presented the negative correlation of radon concentration with outside temperature. However, the positive correlation could be also observed. There was a correlation of radon concentration with the sizes of atmospheric pressure amplitudes observed in the month's period.

The coefficients of seasonal radon concentration changeability determined for the examined buildings approximate the annual value of radon concentration based on 3- or 6-months' exposure. They can be used to assess the mean concentration with reference to larger groups of houses. Such assessments, unlike short-term exposure, represent the risk of population connected with radon inhalation. There cannot be drawn any conclusions, based on determined coefficients, concerning radon concentration changeability in a given building as, in spite of dominating tendencies, particular buildings reveal individual changeability schemes.



# **INTERCOMPARISON OF INSTRUMENTS FOR MEASURING RADON AND RADON PROGENY HELD IN THE CLOR CALIBRATION CHAMBER\***

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Intercomparisons among radon laboratories are of particular importance because calibrated standards for radon activity concentration and for radon progeny activity concentration in air are not available.

An intercomparison exercise for radon and radon progeny instruments was conducted at the Central Laboratory for Radiological Protection (CLOR) from June 9

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\* This work was partially supported by Radon Center.



to 13, 2003 to verify comparability of mainly continuously measuring instruments for radon and potential alpha energy concentration (PAEC). Seven Polish institutions participated to intercompare ten radon and six PAEC monitors. Among radon devices there were: six Genitron AlphaGUARD monitors, two Pylon AB-5 monitors, radiometer RMR1 and a radon probe SRDN2. Among PAEC devices there were five monitors measuring in continuous mode: three T/N Rn WL Meters co-working with AlphaGUARD monitors, Pylon WLx monitor and Radon Progeny Particles Size Spectrometer (RPPSS), and one grab sampling device: aspirator SKC equipped with ALFA unit.

All the instruments were simultaneously exposed in the CLOR big calibration chamber of the volume of 12.35 m<sup>3</sup>, in the radon atmosphere in the range from ca. 2500 Bq/m<sup>3</sup> to ca. 700 Bq/m<sup>3</sup> under controlled different climatic and aerosol conditions. They were changed from normal (45–50%) to very high relative humidity (95–99%) and in both values of the relative humidity from low ca. 200 CN/cm<sup>3</sup> to high concentrations of condensation nuclei ca. 31 000 CN/cm<sup>3</sup>. Aerosols were injected from the TSI water generator. The total exposure lasted ca. 44 hours. A value of radon concentration calculated on the basis of the activity of radium <sup>226</sup>Ra in the certified Pylon flow-through radon generator constituted the reference value for radon at the beginning of the exposure. Later the average results in four periods of time from all instruments could be compared only among themselves due to the loss of radon while walking in and out the chamber.

In the analysis of the PAEC results the spectrometer RPPSS was considered reference one because it measures radon progeny particle size distribution and applies it to correct the PAEC value for plateout of small particles upstream in the inlet of the device.

AlphaGUARD monitors were also compared in the atmosphere of toron in the small calibration chamber of volume of 0.32 m<sup>3</sup>. Toron was pumped into the chamber from the certified Pylon flow through toron generator in closed circuit with flow of 1 dcm<sup>3</sup>/min. The monitors worked as well in the diffusion mode as in flow mode with different two flows: 0.5 and 1 dcm<sup>3</sup>/min.

The results of average radon concentrations for all AlphaGUARD monitors indicate very good agreement for the whole exposure time in the big chamber. For six



AlphaGUARD monitors standard deviations of mean values for four selected periods range from 1 to 2.8% while for all monitors they range from 5.5 to 20%. The comparison of the means for AlphaGUARDs in toron chamber, where readings were on the level of below  $100 \text{ Bq/m}^3$  gave significantly worse results.

The comparison of the PAEC results from T/N Rn WL Meters and Pylon WLx monitor with the reference value from RPPSS indicate differences in the calibration factors ranging from 30 to 42% determined in high aerosol conditions (free fraction of 5%) and the losses of PAEC ranging from 38 to 55% possibly due to plateout in the inlets of filter holders in the conditions of low aerosols concentration (free fraction of 62%).



# AEROSOL SAMPLING FOR RADIOLOGICAL PROTECTION: WHICH PARTICLE SIZE AEROSOL SAMPLER TO SELECT?

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Sampling of radioactive aerosols for the purpose of assessing or predicting occupational radiation doses becomes an important issue in the European countries. In particular, the 96/29/Euratom Council Directive specifies that aerosol sampling results can be used for assessing the individual dose when the individual in vivo and/or bioassay methods are not possible or give insufficient results. It precisely is the case for exposures by inhalation to Naturally Occurring Radioactive Materials (NORMs), for which bioassay methods may be associated with high detection limits while in comparison traditional aerosol sampling methods may lead to lower detection limits in terms of dose.

A generic method has been developed to facilitate the identification of the particle size aerosol sampler (following the inhalable, thoracic or respirable convention) to select for minimising the respective biases between the true and estimated expo-



sure and the true and estimated effective dose associated with exposure by inhalation to any radioactive compound.

Calculation based on this method have been applied to various radioactive compounds (of different absorption rates and particle size dispersion characteristics) of the  $^{238}\text{U}$  and  $^{232}\text{Th}$  natural chains. These calculation have shown that:

- for exposure and effective dose estimates, the sampling efficiency of the sampler should be known and corrected for,
- for exposure estimates, an inhalable sampler should be chosen,
- for effective dose estimates, the sampler should be chosen according to the absorption rate of the considered compound, in order to follow as closely as possible the AMAD dependency of the compound's dose coefficients:
  - a thoracic sampler should be chosen for compounds of slow and moderate absorption rate,
  - an inhalable sampler should be chosen for compounds of fast absorption rate,
  - in absence of (precise) information on the particle size characteristics of the ambient aerosol, the following default values should be chosen:  
AMAD = 5  $\mu\text{m}$ , GSD = 2.5.

This paper presents the key elements of the method and the main results of it's application to exposures by inhalation to NORMs.

This work was partially supported by the European Commission DG Research within the framework of the 5th PCRD (SMOPIE project: "Strategies and Methods for Optimisation of Internal Exposures of workers from industrial natural sources").



# A PROTOCOL FOR DETERMINATION OF NATURAL RADIONUCLIDES IN THE GEOTHERMAL WATER

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A routine radiometric analysis of the underground water including the geothermal water is generally limited to the main natural radionuclides. A simplified protocol for the determination of natural radionuclides in that kind of water has been tested.

Activities of the main radionuclides from  $^{238}\text{U}$  and  $^{232}\text{Th}$  series in the Uniejów geothermal water were determined by combining liquid scintillation counting with  $\alpha/\beta$  separation and  $\gamma$  spectrometry methods. The  $^{222}\text{Rn}$  and  $^{226}\text{Ra}$  activities were measured after extraction of radon from 10 ml water samples to 10 ml of Ultima Gold F scintillation cocktail directly in the 22 ml scintillation vials. The samples were counted in a new generation portable liquid scintillation counter, Betascout, without separation of the phases: over the period of 30 days after extraction. The average values of the specific activities were equal to 2.95 and 0.64 Bq/dm<sup>3</sup> for



$^{222}\text{Rn}$  and  $^{226}\text{Ra}$ , respectively. The  $^{210}\text{Po}$  radionuclide before counting was preconcentrated from 1 dm<sup>3</sup> water samples on hydrated manganese oxide and deposited on silver discs. The discs were immersed in the 10 ml of scintillator and their activity was measured also by the same method. The average  $^{210}\text{Po}$  concentration was 0.052 Bq/dm<sup>3</sup>. Activity of the remaining radionuclides was determined by  $\gamma$ -spectrometry after their preconcentration on the hydrated manganese oxides from 10 dm<sup>3</sup> samples. The activities of two radium radionuclides,  $^{224}\text{Ra}$  and  $^{226}\text{Ra}$ , can be calculated from their basic  $\gamma$ -lines, whereas  $^{228}\text{Ra}$  can be determined from its decay product –  $^{228}\text{Ac}$ , and were equal to 0.52, 0.65 and 0.58 Bq/dm<sup>3</sup>, respectively. The activities of  $^{210}\text{Pb}$  and  $^{238}\text{U}$  ( $^{234}\text{Th}$ ) were below the detection limit of the method equal to 0.03 Bq/dm<sup>3</sup>. Based upon the obtained results, it can be concluded that there are not any radiological restrictions for using this water as a heat source or for balneological purposes. However, it cannot be used as a mineral drinking water, because the calculated committed effective dose from its one year consumption exceeds the WHO recommended value of 0.1 mSv.





# DETERMINATION OF RADIUM $^{226}\text{Ra}$ NUCLIDE IN GROUND-LEVEL AIR

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Installation of the Aerosol Sampling Station (ASS) network in Poland offers the unique opportunity for measuring natural radionuclide concentration in ground – level air. The ASS-500 collects air aerosols on a filter placed 2 m above the ground with the air flow about 500 m<sup>3</sup> per hour in a weekly cycle and each weekly filter is submitted for further  $\gamma$ -spectrometry measurements. Unfortunately, because of slightly higher background levels caused by lead shields in the regions of  $^{234}\text{Th}$  ( $^{238}\text{U}$ ) and  $^{226}\text{Ra}$   $\gamma$ -lines, an achieved detection limits for direct determination of these radionuclides are above the usually occurring concentrations ~1–5  $\mu\text{Bq}/\text{m}^3$ . However, in the case of  $^{226}\text{Ra}$ , such measurements can be possible by recounting of the closed (sealed) filters after a one month period, necessary for establishing of radioactive equilibrium with  $^{222}\text{Rn}$  and its decay products, particularly  $\gamma$ -emitting radionuclides:  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ . The calculated detection and determination limits (with 10% relative accuracy), according to Currie's formulas for these two radionuclides for 160 000 s counting time were: 0.4 and 1.5  $\mu\text{Bq}/\text{m}^3$ , respectively. The observed  $^{226}\text{Ra}$  activities during the half year collection period, including parts of winter and summer seasons, were always above the detection limit and ranged from 0.5 to 3  $\mu\text{Bq}/\text{m}^3$ . The radium radionuclides are present in ground-lev-





# ENVIRONMENTAL RADIOLOGICAL MONITORING IN A NIOBIUM MINING REGION IN BRAZIL

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Mining activities represent a potential source of increment of radiation doses to which the public are exposed (TENORM). In this paper, a niobium mining plant in Brazil is approached in respect to its environmental radiological impact. Radionuclides from the uranium and thorium decay chains occur in the ore, and some of them are concentrated during the metallurgic process. Therefore, an environmental monitoring program was established on a routine basis. The aquatic environment in the influence area of the plant was assessed by measuring samples of drinking water, groundwater, river water, and sediment from riverbed. Samples of atmospheric air were also collected. In the water samples,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{210}\text{Pb}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  were determined by total alpha and beta counting after radiochemical separation and neutron activation analysis. In the sediment samples,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  were determined by gamma spectrometry. In the atmospheric air,  $^{222}\text{Rn}$  and



its decay products were determined by grab sampling. The external exposure was evaluated by thermoluminescent dosimetry. For dose calculations, the pathways taken into account were external exposure and internal exposure, from water consumption and air inhalation. The results obtained in this study show that the mining activities do not imply in any detectable increment to the radiation doses to which individual members of the general public are exposed.



# RADIONUCLIDES IN SWEDISH DRINKING WATER

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Directives from the European Union state that drinking water should be consumed without risk. For radon and other radionuclides in drinking water there are guidelines and recommendations. The EU preparatory working group 1 "Chemical parameters" suggest that a toxicologically based standard should be introduced for uranium. The WHO suggests a guideline value for uranium based on its chemical toxicity to 9 µg/l.

In accordance with the European union the Swedish health authorities have decided on new regulations for drinking water from the 25 of December 2003. The regulations state that the effective dose from radioactivity in drinking water should not exceed 0.1 mSv/year. In this neither tritium, potassium <sup>40</sup>K nor radon and its daughters are included.

The natural radioactivity of Swedish drinking water is high in an international comparison. The reason for this is that the Precambrian bedrock of Sweden, especially granites, are rich in uranium and thorium. Wells drilled in the bedrock can have high concentrations of radionuclides while dug wells generally have very low concentrations.



Central and local authorities have recommended and urged people to analyze radon in their drinking water during the past decades. It is calculated that approximately 35 000 analyses of radon in drilled wells have been performed. From the results it is calculated that 10 000–15 000 out of 200 000 drilled wells in Sweden have radon levels above 1 000 Bq/l.

Analyses of the other radionuclides in drinking water are as yet few. Analyses of radium in 499 drilled wells 1988 showed that radium rarely occur in high concentrations. A former study of uranium in drinking water from 269 drilled wells in Uppsala county gave a mean of 21.1 µg/l and a median value of 12 µg/l.

This study will present results from analyses of radionuclides in drinking water from selected areas of Sweden. In 103 drilled wells was the median content of uranium  $^{238}\text{U}$  7.3 µg/l, for radium  $^{226}\text{Ra}$  the median content was 0.02 Bq/l and for radon the median content was 360 Bq/l. The median value for gross alpha was 0.66 Bq/l and för gross beta 0.3 Bq/l. The correlation between the radionuclides were very weak.



# RELEASE OF NORM-RESIDUES FROM SURVEILLANCE IN GERMANY – SOME PRACTICAL EXPERIENCE

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In Annex XII Part A of the German Radiation Protection Ordinance (StrlSchV) radioactive residues resulting from work activities are specified. If their specific activity is higher than a surveillance threshold (Überwachungsgrenze) they have to be kept under surveillance. The disposal of such residues needs an official notification of clearance. The procedure for obtaining such a notification of clearance requires to file an application by the owner of the residues with the following content:

- declaration of the residues (type, mass, specific activity),
- statement of the disposal company regarding the acceptance of radioactive residues,
- evidence of conformity with the waste law,
- proof that the dose limit of 1 mSv/year to the individual will be kept.

In order to simplify the preparation of the application a well structured document containing all necessary statements and proofs was developed by us. It consists of 6 application formulars and 2 or 3 reports. It enables an effective preparation of



the application form and a speedy examination of the proposal by the authority. However, the first experience obtained show several open problems to be solved in future. These will be presented and discussed in the paper.





# IDENTIFICATION OF ENHANCED CONCENTRATIONS OF $^{210}\text{Pb}$ AND $^{210}\text{Po}$ IN DUST SAMPLES FROM STEEL-WORKS

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A study has been initiated to investigate the naturally occurring radioactivity in byproducts of the industrial iron-ore reduction process such as sinter-dust and blast-furnace dust. In a first step, dust samples obtained from the air stream cleaning procedure at one production place have been investigated. Enhanced concentrations of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  radionuclides have been found through measurements of the emitted gamma and alpha radiations.

Thus, these materials have to be handled according to the regulations outlined in the German Radiation Protection Ordinance, Part 3, Appendix XII, i.e., recycling or disposal of these materials are controlled by law. In the presentation, the measurements performed, the results as well as the specific regulations which have to be applied to these materials will be given.



# TEST OF THE MATERIAL FOR RADON SEAL LAYER AT THE MINE WASTE DISPOSAL SITE JAZBEC

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About 1.4 million ton of the mine waste material was deposited at the mine waste disposal site Jazbec during the uranium ore mining in the uranium mine Zirovski Vrh. The average concentration of uranium in the mine waste is about 60 g/ton, the average specific activity of Radium  $^{226}\text{Ra}$  is  $730 \text{ Bq/m}^3$ . To reduce the radon exhalation rate and to reduce the penetration of the rainfall water into the waste pile it should be covered with an appropriate material. The surface of the waste pile is about 5 ha. The thickness of the cover should be about 2 m.

A test field on the waste pile Jazbec (100 m long and 12–20 m wide) was constructed in the year 2003. The surface of the test field was separated in two parts: one part with multi layer cover (5 layers), the second part with only two layers. For the seal layer the fine clayey silt and sand was used. The measurements of radon exhalation were done on the mine waste material, on the seal layer, on the protection layer and finally on the humus layer. For radon exhalation measurements a method with charcoal absorbers was applied. The time of exposure was 48 hours. The results of radon exhalation rates assure that clayey silt and sand mixture is adequate material to use for radon barrage. The average results of radon exhalation



tion rates are 50% and more percent lower than obligatory authorized limit of 0.1 Bq/m<sup>2</sup>s. The results of the water penetration measurements are not as low as it is required by regulations. The improvement of clayey silt and sand characteristics with bentonite was also evaluated.



# ESTIMATION OF RADON DOSE IN SEVERAL WORKPLACES USING DOSIMETRIC MODEL FOR INHALATION OF AIRBORNE RADIONUCLIDES

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There are two approaches to the estimation of the effective dose associated with the inhalation of  $^{222}\text{Rn}$  short-lived progeny. One of them based on the Publication 65 ICRP (International Commission on Radiological Protection ) using a single dose conversion factor, called conversion convention, is derived from the results of the epidemiological studies of uranium miners. However, the extrapolation from the lung cancer risk for uranium miners applied to the general population meets many objections related to both: big difference of about 2-3 orders of magnitude in the radon exposure, and differences in the exposure environments (e.g. concentration, size distribution and chemical composition of aerosols), breathing rates, smoking pattern and so on. The second approach is based on the Publication 66 ICRP which strongly recommends the usage of dosimetric models for inhalation of airborne radionuclides. These models reveal that the dose per unit intake of radon progeny depends on the site of particle deposition in the respiratory track, which, in turn, strongly depends on the particle size distribution. It is particularly important



in the estimation of the dose to take into account the contribution of the ultrafine particles below 10 nm in diameter. To recapitulate, for the reliable estimation of the radon dose it is necessary to know not only the alpha potential energy concentration (PAEC), but also the full size distribution of radon progeny particles in the range from 1 to 1000 nm, which penetrate to our lungs with the air. The dosimetric model approach uses a weighted dose conversion factor, which combines radon progeny size distribution with the particle-size dependent dose conversion factors in a particular exposure location.

By means of our Radon Progeny Particle Size Spectrometer, manufactured by dr Stephen Solomon in ARPANSA, Melbourne, Australia, we measured potential alpha energy concentration (PAEC) and radon progeny size distribution in five workplaces. They were: an attorney office, Faculty of Physics at the Warsaw University – two locations, the Central Laboratory for Radiological Protection and an experimental coal mine “Barbara”. The measurements were conducted in various conditions of aerosols: natural and high level aerosols from smoking cigarettes. We applied both approaches for the estimation of the annual effective doses from inhaled radon progeny for an adult male with a breathing rates of 1.2 m<sup>3</sup>/h and 0.78 m<sup>3</sup>/h, appropriate respectively to occupational and environmental exposure, and compared them. Ratios of the doses estimated by the dosimetric model to ones with the use of the constant conversion convention range from 0.5 when the free fraction is ca. 5% to 1.7 when the free fraction is ca. 30%.



# **SAMPLE CLEAN-UP BY ON-LINE CHROMATOGRAPHY FOR THE DETERMINATION OF PU AND AM IN SEDIMENTS**

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Plutonium is the most investigated actinide element in samples of different matrices. A complete inventory of its isotopic composition can be obtained by different analytical techniques i.e. mass spectrometry for the determination of  $^{239}\text{Pu}/^{240}\text{Pu}$  isotopic ratio, alpha spectrometry for the  $^{238}\text{Pu}$  and  $^{239(40)}\text{Pu}$  determination, and liquid scintillation counting for the determination of  $^{241}\text{Pu}$ . To obtain good results, separation of Pu from the matrix as well as other actinide elements is necessary. In this paper a novel clean-up procedure for the determination of Pu and, consequently, Am in environmental samples analysed by alpha spectrometry is presented. The method is based on the purification of the samples from different standard matrices (sediments etc.) and separation of Pu and Am by using three different chromatographic columns and two different eluents. The eluted fractions are collected at different times, and are checked for purity by alpha spectrometry.



This procedure, discussed in terms of analytical figures of merit, has been validated with certified reference materials and compared with the classical routine methods for alpha source preparation.



# THE INVENTORY OF ITALIAN NORM CONCERNED WORK ACTIVITIES IN THE FRAME OF ENVIRONMENT PROTECTION

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The importance of NORM in Italy, from the radiation exposure point of view both for workers and for the public, is confirmed by a national decree (n. 241/2000), implementing the EU BSS, that provides the execution of controls for a set of work activities.

A project aimed at estimating environmental impact for some activities dealing with NORM is being carried out by the National Topic Centre on Physical Agents (CTN-AGF), coordination of Regional Environmental Protection Agencies (ARPA), which supports the National Environmental Protection Agency (APAT) in collecting environmental information about physical pollutants (ionizing and non-ionizing radiations, noise). By now the following working categories have been chosen: phosphate and fertilizers industry, integrated steelworks, processing of zircon sands, oil and gas extraction, uranium mines, coal-fired power plants.

Information about presence and features of different working cycles has been obtained through field reports, representatives of sector associations or of the companies themselves.

Radiological surveys have been carried out to increase knowledge for some work activities aspects potentially relevant as far as environmental impact is concerned (on dusts and residues in refractory, tiles industry and in integrated steelworks etc.)

Technical data related to the various work activities have been collected by means of specific questionnaires sent to most of the involved factories: a centralized data base has been set up. An estimate of Italian work activities general environmental pressure, first step for radiological impact predictions, is possible based on both radiological information (present and already available) and technical information from the questionnaires.



# URANIUM ISOTOPES IN PUBLIC DRINKING WATER IN POLAND

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$^{238}\text{U}$ ,  $^{234}\text{U}$  and  $^{235}\text{U}$  were determined in tap water samples collected from water supply systems located in various parts of Poland. The systems were supplied with water from rivers or lakes (surface water) and from underground wells (underground water).

Uranium isotopes were determined in 5-L samples.  $^{232}\text{U}$  was used as an internal tracer. After water evaporation, uranium was chemically separated by anion exchange and extraction. Finally uranium was electrodeposited on a stainless steel disc and activity of uranium isotopes was measured by alpha spectrometry.

Average activity concentrations of  $^{238}\text{U}$ ,  $^{234}\text{U}$  and  $^{235}\text{U}$  in tap waters originating from surface waters were  $10.3\pm 1.96$  mBq L<sup>-1</sup>,  $13.8\pm 2.68$  mBq L<sup>-1</sup> and  $0.44\pm 0.08$  mBq L<sup>-1</sup>, respectively. Average activity concentrations of these isotopes in tap water originating from underground waters for  $^{238}\text{U}$  and  $^{234}\text{U}$  they were  $4.53\pm 1.50$  mBq L<sup>-1</sup> and  $5.68\pm 1.72$  mBq L<sup>-1</sup>, respectively, whereas for  $^{235}\text{U}$  in six out of 16 water pipes the concentrations were below the lower limit of detection (0.06 mBq L<sup>-1</sup>), and for the other the average was  $0.29\pm 0.09$  mBq L<sup>-1</sup>.



Activity concentrations of  $^{234}\text{U}$  were higher than those of  $^{238}\text{U}$  (atomic  $^{234}\text{U}/^{238}\text{U}$  ratios ranged from 1.07 to 2.6), indicating the lack of equilibrium between these isotopes. The  $^{234}\text{U}/^{238}\text{U}$  ratios for the surface and underground waters were similar.

The average activity ratio of  $^{235}\text{U}$  to  $^{238}\text{U}$  for water samples was equal to  $0.045 \pm 0.003$  which is close to the value 0.046 for uranium in nature.

In seven water supply systems the samples were taken before and after water treatment. Only in one water source the treatment reduced the uranium content significantly (by about a half), while in the other sources the treatment usually did not decrease the uranium content. The activity concentrations of uranium isotopes in tap waters taken from surface waters were similar to those from rivers and lakes in Poland [1].

#### Reference

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# CHARACTERISATION OF SCALE FROM A FORMER PHOSPHORIC ACID PROCESSING PLANT

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A wide variety of industries extract and/or process ores and minerals containing naturally occurring radioactive material (NORM). In many cases the industrial processing of natural feedstock results in radioactive material becoming concentrated in (by-) products, wastes and plant installations. The main sources of NORM are members of the  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay chains. The radioelements of greatest concern are radium, lead and polonium, due to their radiological and toxicological properties when inhaled and/or ingested (Kathren, 1998). Therefore, it is necessary for radioactive materials to be isolated from the human environment, thereby committing industries to costly disposal initiatives under the current directives.

Scale samples were collected from a phosphoric acid processing plant, which is currently in the process of being demolished. These materials were analysed



chemically, mineralogically and radioactively to determine the nature and concentration of the radionuclides retained in pipe work and process plant (Poole et al., 1995). Three areas of the site were investigated; 1) the Green Acid Plant, where crude acid was concentrated; 2) the green acid storage tanks; and 3) the Purified White Acid (PWA) plant, where inorganic impurities were removed. The scale samples predominantly comprised: fluorides (e.g. malladrite, iron oxide fluoride); calcium sulphate (e.g. bassanite); and an assemblage of fluorides and phosphates (e.g. iron fluoride hydrate, calcium phosphate) respectively, which can be related to the process occurring at each of these locations. The radioactive inventory is primarily all from  $^{238}\text{U}$  and its decay chain products. Compared to the feed-stock ore values, elevated concentrations, to levels  $\leq 8.8$  Bq/g of  $^{238}\text{U}$ , and its daughter  $^{234}\text{Pa}$ , were found to be retained in installations where the process stream was rich in fluorides and phosphates. Enriched levels,  $\leq 11$  Bq/g of  $^{226}\text{Ra}$  were found in association with precipitates of calcium sulphate. Fractionation of  $^{226}\text{Ra}$  from its daughter  $^{210}\text{Pb}$  was observed in some scales; however,  $^{210}\text{Pb}$  levels in excess of its parent was also noted in many materials from the PWA plant, which may be attributed to the purification process occurring in this installation. In addition, enriched concentrations of trace elements Pb ( $\leq 106$ ), Cu ( $\leq 152$  ppm), Ni ( $\leq 215$  ppm), Zn ( $\leq 2183$ ) and particularly Cr ( $\leq 3110$  ppm) were also found in many of the materials analysed.

Reference:

Kathren R. L. (1998) NORM Sources and Their Origins. *Applied Radiation and Isotopes* 49(3), 149-168.

Poole A.J., Allington D.J., Baxter A.J., and Young A.K. (1995) The natural radioactivity of phosphate ore and associated waste products discharged into the eastern Irish Sea from a phosphoric acid production plant. *The Science of the Total Environment* 173/174, 137-149.



# VENTILATION: A TECHNIQUE FOR INDOOR RADON MITIGATION

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The problem of radon is an important global problem of radiation hygiene concerning the world population. It has been estimated that the radon, largely in homes, constitutes more than 50% of the dose equivalent received by general population from all sources of radiation, both naturally occurring and man-made. When radon decays to form its progeny ( $^{218}\text{Po}$  and  $^{214}\text{Po}$ ), they are electrically charged and can attach themselves to tiny dust particles, water vapours, oxygen, trace gases in indoor air and other solid surfaces. These daughter products remain air borne for a long time. These dust particles (aerosols) can easily be inhaled into the lung and can adhere to the epithelial lining of the lung, thereby irradiating the tissue. As per the ICRP recommendations, it is essential to adopt remedial measures if the radon levels in dwellings are found to be more than  $200 \text{ Bq m}^{-3}$ . Ventilation process can simulate the conditions generated through advection or diffusion and can be effective in mitigating indoor radon.



In the present work, the effect of ventilation (natural and forced by exhaust fan used at different speeds), on radon concentration in a room having an external source of radon was studied. For radon, the LR-115, type II solid state nuclear track detectors (SSNTDs) were used. The radon reduction factor, which is the ratio of radon concentrations before and after remediation was calculated. The radon reduction factor was found to vary from 1.08 to 1.17 due to natural ventilation whereas 1.17 to 3.01 due to forced ventilation. The results indicate that optimized ventilation (natural or forced) can be a simple mean to mitigate radon in dwellings.

Key words: Radon concentration, remediation, ventilation, SSNTDs, exhaust fan

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# **IN SITU GAMMA-RAY SPECTROMETRY IN COMMON ROCK RAW MATERIALS MINED IN KRAKOW VICINITY, POLAND**

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The natural radioactivity of  $^{40}\text{K}$ ,  $^{208}\text{Tl}$ ,  $^{212}\text{Pb}$ ,  $^{212}\text{Bi}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ ,  $^{228}\text{Ac}$  and the fallout of  $^{137}\text{Cs}$  in common rock raw materials mined in Krakow vicinity were measured in situ using a portable gamma-ray spectrometry workstation. The measurement points were chosen for different regional rocks in: Devonian and Jurassic limestones, dolomite, trachyandesite, porphyry, porphyry tuff, diabase and melaphyre.  $^{40}\text{K}$  activity varied in the range from about  $82 \text{ Bqkg}^{-1}$  (Jurassic limestone) to  $3150 \text{ Bqkg}^{-1}$  (porphyry tuff). The activity concentrations associated with  $^{228}\text{Ac}$  ( $^{232}\text{Th}$  series) varied in the range from  $9 \text{ Bqkg}^{-1}$  (Jurassic limestone) to  $56 \text{ Bqkg}^{-1}$  (porphyry tuff), whereas activity of  $^{226}\text{Ra}$  ( $^{238}\text{U}$  series) ranged from about  $13 \text{ Bqkg}^{-1}$  (Jurassic limestone) to  $43 \text{ Bqkg}^{-1}$  (Devonian limestone). The highest deposition of  $^{137}\text{Cs}$  (20





kBqm<sup>-2</sup>) was recorded in Devonian limestone. Since many of these rocks are used as building materials, evaluation of radioactive elements concentrations in these materials is important because indoor radiation exposure can vary depending on used building materials.



# SOURCES OF TENORM – INVENTORY OF PHOSPHATE FERTILIZERS AND ALUMINUM INDUSTRIES

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## I. Phosphate fertilizers industry

In Romania, there were built and commissioned in the period 1964–1978, 7 fertilizers plants based on  $H_2SO_4$  and  $HNO_3$  attack.

### A $H_2SO_4$ attack

The deposit with sub-products (phosphogypsum) from SC SOFRET SA Bacau. The material (phosphogypsum) resulted from  $H_2SO_4$  attack of the imported phosphatic rocks has been deposited since 1978, close to the plant, which is located in the Eastern part of Bacau city. In the samples resulted from the surface of the phosphogypsum deposit and also from drills until a depth of 16 m there were detected the following contents in radioelements: U = 15–21 ppm,  $^{226}Ra$  = 0.45–0.87 Bq/g, Th = <3 ppm, K = <1%, gamma dose flow rate between 0.18–0.25  $\mu Sv/h$ . Radon concentration at the pond surface (irregular areas) was of 4860 Bq/m<sup>3</sup> and for thoron was of 496 Bq/m<sup>3</sup>.



### B HNO<sub>3</sub> attack

The tailing pond with sub-products from S.C. AZOMURES S.A. Targu-Mures. The tailing pond with technological water and calcium carbonate occupies a surface of about 33 ha, close to the plant, which is located in the Western part of Targu-Mures city. The technological water from the tailing pond has a pH = 1.45 and the contents in U = 0.045 mg/l and Ra = 1.4 Bq/l. The sub-product (calcium carbonate) contains 23 ppm U, 0.35 Bq/g Ra, 2 ppm Th and 0.2% K.

### II. Aluminum industry

#### A. Alumina Plant from Oradea

In the western area of Oradea city there is the bauxite processing plant. In the pond with red sludge resulted from alumina acquirement, the contents in radionuclides are mentioned below (Table 1):

Table 1

	ppm	Bq/g	ppm
	U	Ra	Th
Material of red colour	11	0,21	61
Material of dark colour	<10	0,137	70
Material of light colour	127	1,675	11

In air at the pond surface: <sup>222</sup>Rn = 21 Bq/m<sup>3</sup>, thoron = 1220 Bq/m<sup>3</sup>. In water from the red sludge pond: <sup>222</sup>Rn = 101 pCi/l, thoron = 0, <sup>226</sup>Ra = 0.023 Bq/l, pH = 12.05.

#### B. Alumina Plant from Tulcea

Sub-product – red sludge: U = 15 ppm; <sup>226</sup>Ra = 0.212 Bq/g; Th = 62 ppm. In water from pond: pH = 12.2; <sup>226</sup>Ra = 0.026 Bq/l; U = 0.010 mg/l. <sup>222</sup>Rn and thoron distribution in air at the red sludge pond surface: <sup>222</sup>Rn = 248 Bq/m<sup>3</sup>, thoron = 6270 Bq/m<sup>3</sup>.



# **RADON MEASUREMENTS AS A MONITORING POSSIBILITY FOR MINING SUBSIDENCE OCCURRENCES**

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The objective of the current research is to check whether or not radon concentrations in the surface layer can be related to mining and mining subsidence voids in order to allow the drawing of a subsidence risk map.

The chosen test site is located in a forested area on the plateau of the Luxembourg mining basin. Under an overburden of 65 m thick limestone and marls two approximately 3-m-high iron layers have been worked between 1920 and 1950. Due to the prevailing room-and-pillar extraction method, pillars of different shape and size have been kept in place. Mining subsidence has progressively occurred for more than 50 years and is currently going on. Along the western mine boundary a long surface fissure has developed and some 25 subsidence pits occurred over a period of several decades, the last in 1995. The site was mainly chosen because it has a more or less uniform surface, the old mining maps still exist and fur-



ther subsidence may be expected the more than the neighbouring busy road has not been protected by a continuous pillar.

The ease of radon to leave the site of production in the soil and rocks and the differences in the subsequent possibility to migrate and concentrate in underground voids, give the possibility to locate underground inhomogeneities by a survey based on radon soil measurements. Normally mining voids situated at a depth of 60 m have no influence on radon concentrations measured in one meter depth. But due to mining subsidence, an important brittleness of the rocks can develop moving up to the surface; as a consequence we expect locally increased radon concentrations. We report of an intensive investigation on radon in soil measurements, distributed over 10 parallel transects. Along a transect the spacing between the measurement points was 4 meter, for 8 transects the inter-distance was 10 m, it was 30 m for the last two transects.

Radon concentrations plotted against sample spacing are very irregular documenting a very inhomogeneous subsoil. The map of the radon concentrations (Fig.1) shows these irregularities, but on the whole it is possible to identify a trend from East to West, corresponding to the main directions of the old mine roadways. The location of enhanced radon concentrations may present an increased subsidence risk consecutive to a fractioning of the underground. This study documents that investigations based on radon in soil can contribute to risk investigation related to mining subsidence occurrences.

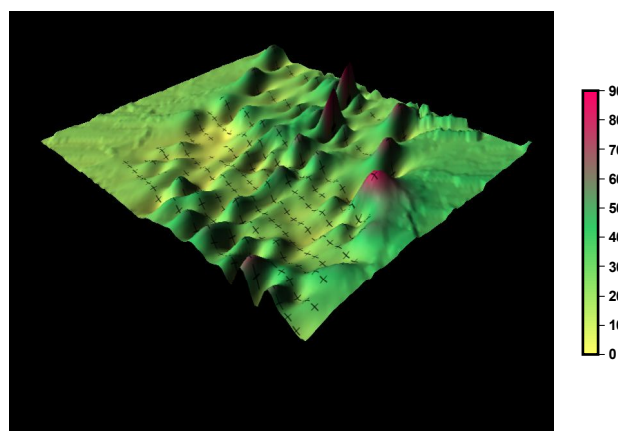


Fig. 1: Map of radon concentrations (in Bq/m<sup>3</sup>)



# NATURALLY OCCURRING RADIOACTIVE MATERIAL (NORM) ASSESSMENT OF OIL AND GAS PRODUCTION INSTALLATIONS IN NIGERIA

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Naturally Occurring Radioactive Material (NORM) assessment was carried out at oil production platforms and gas processing units in Nigeria with the aim of establishing baseline levels and problem areas. The assessment consisted of external radiation measurements on production units, from the wellheads to the product outlets, sampling and gamma spectrometric analysis of scales and sludge from pig stations and of replaced pipes and vessels. The NORM levels on the installations and associated equipment ranged from 0.1 to 15  $\mu\text{Sv/h}$  and  $\gamma$  spectrometric analysis indicate the presence of  $^{226}\text{Ra}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Pb}$  in the scales and sludge having maximum concentration of 200 Bq/g. The levels may seem low but there is still the need to establish routine monitoring program for the Industry, which presently is non-existent.





# **NATURALLY OCCURRING RADIONUCLIDES IN RAW MATERIALS, PRODUCTS AND WASTE IN THE STEEL INDUSTRY**

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The Institute for Ferrous Metallurgy (IFM) activities in the field of the monitoring of the radioactive contamination in the steel industry are presented. The research was carried out in the framework of EUREKA “Intracem” project.

The aim of the activities is development of integrated radiation protection system for Polish steel industry (comprising staff, products, waste and environment). Definition of “steel industry” includes also other cooperating manufactures and companies i.e.: suppliers or producers of scrap, raw materials and refractory products.

The scope of interest includes both artificially and naturally occurring contaminating radionuclides (NORM).





# CHARACTERISATION OF SCALE FROM A FORMER PHOSPHORIC ACID PROCESSING PLANT

COMPLETE LECTURE TEXT

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A wide variety of industries extract and/or process ores and minerals containing naturally occurring radioactive material (NORM). In many cases the industrial processing of natural feedstock results in radioactive material becoming concentrated in (by-) products, wastes and plant installations. The main sources of NORM are members of the  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay chains. The radioelements of greatest concern are radium, lead and polonium, due to their radiological and toxicological properties when inhaled and/or ingested (Kathren, 1998). Therefore, it is necessary for radioactive materials to be isolated from the human environment, thereby committing industries to costly disposal initiatives under the current directives.

Scale samples were collected from a phosphoric acid processing plant, which is currently in the process of being demolished. These materials were analysed



the bed refractory masses). There were carried out the analysis of the possibility of the implementation of contaminated mould fluxes as a tracers of the surface non-metallic inclusions in steels.



# ANALYTICAL METHODS USED FOR DETERMINING NATURAL RADIONUCLIDES IN FINNISH DRINKING WATER

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Natural radioactivity in Finnish drinking water sources comes mainly from decay series of  $^{238}\text{U}$ . Radon ( $^{222}\text{Rn}$ ) has the greatest contribution to the radiation dose. The long-lived alpha-active isotopes of uranium ( $^{238}\text{U}$  and  $^{234}\text{U}$ ), polonium ( $^{210}\text{Po}$ ) and radium ( $^{226}\text{Ra}$ ) and beta-active lead ( $^{210}\text{Pb}$ ) and radium ( $^{228}\text{Ra}$  from  $^{232}\text{Th}$  series) isotopes cause high doses very rarely. For most of Finns, the radiation dose caused by natural radionuclides in drinking water is low. Only those people, who consume water from drilled wells, may obtain high doses via drinking water. During the last 35 years STUK has studied radioactivity in drinking water that originate either from surface or ground water sources. Now more than 1.000 waterworks, 8.000 wells drilled in bedrock and 5.000 wells dug in soil have been studied. The present database includes results from 50.000 measurements or analyses.

$^{222}\text{Rn}$  is measured with a liquid scintillation spectrometer in a homogeneous solution. Most samples coming for radon measurement are screened by gross alpha and beta measurements in order to find the occurrence of  $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{226}\text{Ra}$ ,



$^{210}\text{Po}$ ,  $^{40}\text{K}$ ,  $^{210}\text{Pb}$  and  $^{228}\text{Ra}$  in water. Uranium ( $^{238}\text{U}$ ,  $^{234}\text{U}$ ) and  $^{226}\text{Ra}$  contents can be calculated from gross alpha spectrum fairly accurately for most of the samples. Nuclide specific analysis with radiochemical methods are carried out if more precise information of  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{210}\text{Po}$  or  $^{210}\text{Pb}$  contents are required.

Measurements are nowadays carried out with the liquid scintillation spectrometers, Quantulus 1220TM (for gross alpha and beta,  $^{210}\text{Pb}$ ) and Guardian 1414TM (for radon and gross alpha) and with the alpha spectrometer, AlphaAnalystTM (for uranium and  $^{210}\text{Pb}$ – $^{210}\text{Po}$ ).



# **RADIUM LEACHING FROM MINE DEPOSITS AS A POSSIBLE SOURCE OF GROUNDWATER CONTAMINATION**

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Saline waters occurring in underground coal mines in Poland often contain natural radioactive isotopes, mainly  $^{226}\text{Ra}$  from uranium series and  $^{228}\text{Ra}$  from thorium series. Approximately 40% of total amount of radium remains underground in a form of radioactive deposits, but 225 MBq of  $^{226}\text{Ra}$  and 400 MBq of  $^{228}\text{Ra}$  are released daily to the rivers with mine effluents through surface settling ponds. Very peculiar situation is observed in coal mines, where as a result of precipitation of radium from radium-bearing waters radioactive deposits are formed. Sometimes natural radioactivity of such materials is very high, in case of scaling from coal mines radium concentration may reach 400 000 Bq/kg – similar activity as for 3% uranium ore. Usually such deposits can be found underground, but sometimes co-precipitation of radium and barium takes place on the surface, in settling pond and in rivers. Therefore maintenance of solid and liquid waste with technologically enhanced natural radioactivity (TENORM) is a very important subject.



Lately another problem appeared – due to the decrease of the production of Polish coal industry and dismantling of several coal mines, also the ground reclamation should be done in their vicinity. But in several cases deposits in the ponds contain enhanced levels of radium concentration. Therefore laboratory tests were done to investigate a possibility of the re-entry of radium into groundwater or river waters from such deposits. Results show, that in case of insoluble barium and radium sulphates co-precipitated out from waters type A, re-entry ratio is very small. Different situation can be observed in case of radium, adsorbed on bottom sediments from waters type B, because re-entry ratio is much higher. Nevertheless, this phenomenon seems to be not so important and significant for the further pollution of the adjacent areas of the settling ponds in the future.



# MEASUREMENT OF SHORT-LIVED RADON DAUGHTERS IN POLISH MINES

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Obligatory measurements of the potential alpha energy concentration of short-lived radon daughters have been performing in the Polish underground mines since 1989 year. In consideration of economical aspects, it is attempted from the very beginning to combine it with measurements of the dust concentration. Therefore the developed measuring units were an integral part of the dust samplers complying the requirements of the State Mining Authority to apply it in underground mines. This way the developed devices could fulfil two measurement tasks simultaneously: measurement of the dust concentration and potential alpha energy concentration of short-lived radon daughters. The new device based on the thermoluminescence detectors is able to co-operate with the SKC universal pumps equipped with a cyclone making it possible to operate constantly for a one working day. The lower limit of detection was equal about  $0.04 \mu\text{J}/\text{m}^3$  at a 95% confidence level and 1-hour pumping.



# RADIUM REMOVAL FROM MINE WATERS – UNDERGROUND TREATMENT INSTALLATION

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In underground coal mines in the Upper Silesian Coal Basin there are inflows of highly mineralised waters containing radium isotopes. These waters cause radioactive pollution of the natural environment in mining areas. Therefore cleaning of saline waters of radium is very important. Two types of radium-bearing waters were distinguished – one type containing radium and barium ions, but no sulphates (type A) and another one in which radium and sulphate ions are present but no barium (type B).

A very efficient and inexpensive method of purification of saline waters of  $\text{Ba}^{2+}$  and  $\text{Ra}^{2+}$  ions was developed and implemented in two coal mines. As the result of used technology, based on application of phosphogypsum as the cleaning agent, a significant decrease of radium discharge was achieved – daily of about 120 MBq of  $^{226}\text{Ra}$  and 80 MBq of  $^{228}\text{Ra}$ .

Another type of radium waters does not contain barium ions, but contains sulphate ions  $\text{SO}_4^{2-}$ . There is no carrier for co-precipitation of radium so radium is trans-





ported with discharged waters to main rivers. Different method of purification from radium must be applied for such waters. Laboratory and field experiments were performed, and a cleaning method was chosen. For purification of saline waters – waste products from other industrial processes are applied. The method of purification has been applied in full technical scale in coal mine with very good results – of about 6 m<sup>3</sup>/min of radium-bearing waters is cleaned. Whole this process takes place in underground old workings without any contact of mining crew with radioactive deposits, which are produced during the process. As a result radium amount released to the natural environment was significantly diminished – approximately of about 90 MBq of <sup>226</sup>Ra per day and 150 MBq of <sup>228</sup>Ra.



# RADIUM BALANCE IN DISCHARGE WATERS FROM COAL MINES IN UPPER SILESIA REGION

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Saline waters from underground coal mines in Poland often contain natural radioactive isotopes, mainly  $^{226}\text{Ra}$  from the uranium decay series and  $^{228}\text{Ra}$  from the thorium series. Approximately 60% of the total amount of radium remains underground as radioactive deposits, but 120 MBq of  $^{226}\text{Ra}$  and 200 MBq of  $^{228}\text{Ra}$  are released daily into the rivers along with the other mine effluents from all Polish coal mines. Technical measures such as inducing the precipitation of radium in gobs, decreasing the amount of meteoric inflow water into underground workings etc., have been undertaken in several coal mines, and as a result of these measures the total amount of radium released to the surface waters has diminished by about 60% during the last 5–6 years.

Mine water can have a severe impact on the natural environment, mainly due to its salinity. However associated high levels of radium concentration in river waters, bottom sediments and vegetation have also been observed. Sometimes radium



concentrations in rivers exceed  $0.7 \text{ kBq/m}^3$ , which is the permitted level for waste waters under Polish law. The extensive investigations described here were carried out for all coal mines and on this basis the total radium balance in effluents has been calculated. Measurements in the vicinity of mine settling ponds and in rivers have given us an opportunity to study radium behaviour in river waters and to assess the degree of contamination.



# THEORETICAL STUDY OF RADIUM BEHAVIOUR IN AQUIFERS

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In the paper, a theoretical approach to the problem of radium presence in mineralized mine water, is presented. Two main types of radium-bearing waters have been found in Polish coal mines. In type A waters, radium isotopes are present together with barium, while concentrations of sulphate ions are very low. Additionally, in these waters a ratio of  $^{226}\text{Ra}:^{228}\text{Ra}$  activity is usually higher than 1. In type B waters, no barium can be found, but radium together with sulphate ions. Contrary, in such waters the isotopic ratio of radium  $^{226}\text{Ra}:^{228}\text{Ra}$  is below 1, and activities of both isotopes of radium are lower as in type A waters. No other differences in chemical composition of mine waters have been observed.

Analysis shows, that the activity ratio of radium isotopes is related to the dynamics of radium adsorption on the grains of solid phase in the aquifer. During analysis must be taken into account, that the radium build up in formation water due to recoil effect, is stable in time. Additionally, no correlation with elevated concentrations of uranium and thorium in rocks, have been observed. Therefore the enhanced radium content in formation waters must be caused by its mineralization.



The relatively short half life of  $^{228}\text{Ra}$  (6 years) shows, that the process of radium transfer from solid into liquid phase is a short term process for geological scale. Therefore radium content in mine waters must be related to the concentration of natural radionuclides in the close vicinity of the aquifer or the water reservoir.



# RADIUM BEHAVIOUR DURING DESALINATION PROCESSES OF MINE WATERS

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In 80's and 90's investigations of radium behaviour in desalination plant have been performed by Laboratory of Radiometry of the Central Mining Institute. This plant has been located in Dębieńsko Mine for complete removal of dissolved solids from mineralized mine waters. The installation was a very unique solution, the only one constructed for such purposes in Poland and all over the world. In the plant brines from two coal mines have been treated. These salty waters were additionally radium bearing ones, containing enhanced levels of radium isotopes ( $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ ). Our investigation showed enhanced concentrations of radium in different waste materials and by-products of the process. Therefore the economical use of such products wasn't possible.

In last years a plan of the construction of the desalination plant for another group of mines was under the discussion (Piaś and Ziemowit Collieries). Results of investigations of contamination of the natural environment, caused by radium-bearing waters from these mines, indicated how severe this problem was. To



solve this influence, the removal of radium in underground galleries has been applied in one of these mines.

On the basis of previous investigations in Dębieńsko Desalination Plant, an analysis of radium behaviour in planned installation has been prepared. It was done for two possible scenarios – without and with radium removal from mine waters, preceded the desalination process. Results of the analysis are presented in the paper. This assessment gives a very good opportunity to point out how important may be radium removal from mine waters – not only to minimize environmental pollution but also to enable utilization of certain products of desalination process.



# INVESTIGATIONS OF SURFACE SETTLING POND

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One of the problems, which may appear during ground reclamation of surface settling ponds of underground mines, is enhanced radon exhalation from bottom sediments. This problem would become more important, when concentration of radium isotopes in sediments is enhanced due to discharge of radium-bearing waters into pond. For investigations, special radon accumulation chambers have been designed and constructed. Concentration of radon in these chambers can be measured with application of Lucas cells or activated charcoal detectors. In the latter method radon is extracted from charcoal into liquid scintillator and such sample is measured in liquid scintillation spectrometer. Therefore the lower limit of detection

(LLD)

of the second method is as low as  $0.1 \text{ mBq/m}^2\cdot\text{s}$ . while the LLD for Lucas cells is only  $1.5 \text{ mBq/m}^2\cdot\text{s}$ .

Mentioned above methods of measurements of radon exhalation have been applied for investigations of surface settling pond of one Polish coal mines, abandoned and emptied at the beginning of year 2002. An agreement of the mine's management and the local authority is to make the ground reclamation of the





pond. A thick layer of sediments with enhanced concentration of radium isotopes covers the pond's bottom. Maximum concentration of radium isotopes in these sediments is as high as 2.0 kBq/kg for  $^{226}\text{Ra}$  and respectively up to 4.0 kBq/kg in case of  $^{228}\text{Ra}$ . Two years after the complete release of brines from the pond, bottom sediments are still soaked with water. Therefore measurements of radon in soil gas weren't possible. On the other hand, in some parts of the pond investigations of radon exhalation coefficient were done. Preliminary results of measurements, done in 2002 year, showed that in specific sites of the pond, radon exhalation rates were higher as the highest values of radon exhalation from the ground in Upper Silesia region. Values of exhalation coefficient up to 200 mBq/m<sup>2</sup>·s have been found. It must be pointed out, that preliminary measurements were done in period, when water has been only partly removed from the pond and further dry-up of sediments should lead to the increase of radon exhalation.

Data from the year 2003 confirm this assumption. During this period water content in the bottom sediments has been significantly lower as previously and results of measurements of radon exhalation were sometimes above 300 mBq/m<sup>2</sup>·s.

Vegetation transgression into the pond occurred during year 2003, mainly weeds. Samples of vegetation have been collected from this area to measure radium isotopes content and calculate transfer factors for radium isotopes. Very high concentrations of radium isotopes have been found at several sites. Another reason of the pollution might be the leaching of radium from sediments and contamination of groundwater.

Therefore mentioned above sources of radiation hazard must be taken into account for planning and designing of reclamation operations for surface settling ponds of underground mines.



## RADON IN SOME CAVES OF KRAKOW-CZESTOCHOWA JURASSIC UPLAND

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Radon levels in caves and migration processes have been investigated worldwide from many years. In Poland, contrary, there is no systematic measurements in caves, except Lower Silesia region [3], although radon levels may reach several kBq/m<sup>3</sup> in particular caves – see tabel below.

Cave	<sup>222</sup> Rn (kBq/m <sup>3</sup> )
Bear's Cave (Poland) [3]	1.3
Gyokusen-do (Japan) [3]	1.5
Postojna Cave (Slovenia) [2]	2.3
Mitchelstown Cave (Ireland) [1]	3.1
Altamira (Spain) [3]	3.2

In the papers results of radon investigations in chosen caves of Kraków-Częstochowa Upland are presented. Measurements have been performed during summer of year 2003, from June to September. Different caves have been chosen for our investigations – turistic ones with easy access, and caves with difficult access, where specialistic speleological devices must be used. From geological



point of view, caves located on the Upland were developed in karstic processes in Upper Jurassic limestones.

Maximum radon concentration has been found in Straszycowa Góra Cave – 7.3 kBq/m<sup>3</sup>, while lowest concentrations were measured in Zielona Góra Cave (near Częstochowa) – 0.015 kBq/m<sup>3</sup>. A correlation between geological structure and radon concentration in caves has been found. Usually higher radon concentrations have been measured in caves, where tectonic deformations were negligible and roof rocks weren't weathered and fissured. An example of such type is mentioned earlier Straszycowa Góra Cave, which walls and roofs are cemented by calcite, filling and insulating all fissures.

Opposite situation can be seen in caves, developed along huge fissured faults. Examples of such caverns are Piętrowa Szczelina Cave, Maćiwody Cave, Głęboka Cave or Zielona Góra Cave, where thickness of roof rocks is several meters only, moreover these rocks are strongly weathered, therefore radon concentrations are low.

Till now, no correlation between radon levels and depth of particular caves has been found. Very important problem is to pay special attention to caves with easy access. In the Upland area 5 turistic caves are located and several others with very easy access, but in no caves regular radon monitoring is done.

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# NORM LEGISLATION IN POLAND

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Polish Parliament (Seym) established November 29, 2000 year new Atomic law, changing it significantly many regulations in compliance with EU law. Atomic Law is in force since January 1, 2002 year.

In this act the regulations of peaceful appliance of the atomic energy, real and potential risk due to the ionizing radiation from the artificial sources of radiation or nuclear materials, nuclear wastes and used nuclear fuel. The regulations of the nuclear safety, radioprotection of human and environment has been described in this act, too.

In July 2001 year, Seym changed significantly Polish Geological and mining law. This act is valid since January 1, 2002 year. In this act the regulations for the radioprotection of workers from natural sources of ionizing radiation were significantly modified.

In this paper these new regulations are described with a special emphasis on the lack of detailed regulations as regards of the mining wastes with enhanced natural radioactivity.



# NORM IN MINING INDUSTRY IN POLAND

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The main branches of Polish mining industry are the coal, copper, zinc and lead, salt and oil and gas mines. In many of these mines the problem of the naturally occurred radioactive materials is present.

The main source of risk for the miners is due to the short-lived radon daughter products present in air. This source of risk is present in all Polish underground mines. In coal, copper and oil and gas mines the radium-bearing waters are present. This is a potential risk for the miners, but for the environment too, because all mine water is pumped out to the rivers. In coal and copper mines radium precipitates from the waters creating the risks of the internal contamination but of external gamma radiation too.

The Polish geological and mining law needs to perform monitoring of all sources of natural radiation at the workplaces and to introduce the preventive measures, if necessary. In this paper the results of the monitoring and prevention measures are described.



# THE ASSESSMENT OF EXPOSURE TO IONIZING RADIATION AT SPOIL BANKS

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By the 1970 year Polish hard coal mining was not considered as source of radiation exposure due to presence of enhanced concentration of natural radionuclides. After the radioactive scales in pipelines draining of brines had been found, the necessity of radiation risk's monitoring emerged. During few next years a lot of different surveys have been carried out that resulted in the complex system of occupational radiological protection. In 1982 year due to relevant national rules the system has been enforced into the whole of Polish underground mining. The system consists of systematic measurements of particular factors of the miners' radiation risk, like radioactive aerosols, sediments and waters. Unfortunately, less attention has been given to exposures that can occur as a result of releasing of naturally radioactive materials to environment.

The territory the Upper Silesian Coal Basin is densely populated. Since the phenomenon of enhanced natural radioactivity has become well known to the members of the public a lot of social troubles appear. Majority of them was unexpectedly related to waste rocks piles that are deemed to be as danger as radioactive scales



and sediments. According to claims of local societies the assessment of the radiation exposure at waste rock piles has been done.

Concentrations of natural radionuclides in extracted from underground rocks slightly exceed the natural background on the Earth surface. This fact results in an enhanced gamma dose rate at the places where this kind of waste was dumped. Additionally the structure of spoil banks creates possibility of radon exhalation's increasing. Taking into consideration the total amount of spoil and gangue and the current international recommendation, the enhanced level of external radiation and radon exhalation seems to be significant from point of view of radiological science.

Due to the fact that the waste rock are not undergone any chemical transformation the potential enhanced radiation risk is directly related to geological origin of rocks that constitute that waste. Therefore for investigation two different mining regions have been chosen. The coal seams, exploited in those regions, lie on different carboniferous system so that properties of rocks occurring together with coal differ each other. Measurements of radiation risk have been done on three waste rock piles that vary in age and stage of land reclamation. For comparison the same measurements have been done on neighbouring rural areas. The results of investigations shows that the major contribution to slightly enhanced radiation risk at these areas derives from external irradiation due to enhanced concentration of natural radioactivity in waste material.



# RANKING OF TENORMS IMPACT INTO RADIATION RISK

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After the TENORM phenomenon had been considered thoroughly it turned out that issues occur very frequently in our environment. It touches a lot of aspects of our common life, starting from occupational risk at work places, through some “contaminated” goods or even a visit in spa, and ending on huge amount of bulk waste materials very often dumped in our vicinity. Each particular way of occurring of TENORM determines some unique scenario of exposition. Therefore there is not a universal method for risk evaluation applicable for all possible situations. Moreover, TENORM occurrence’s consequences can be assessed from different points of view. Sometimes the public comprehension of TENORM is a plentiful source of very serious effects far more detrimental and painful than direct exposition to radiation. Risk caused by TENORM is a subject where enforcing of ALARA rule became very complex and multidimensional.

In this paper some proposal of TENORM’s classification and rules of its importance assessment from diverse points of view are described.





# TOWARDS A METHODOLOGY FOR IDENTIFYING NON NUCLEAR SITES WITH ENHANCED LEVELS OF RADIOACTIVITY

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The law of 12<sup>th</sup> December 1997 entrusts ONDRAF / NIRAS, among other things, with the compilation of a repertory, detailing the location and status of all nuclear sites and all sites containing radioactive substances on Belgian territory. This repertory not only includes sites having a nuclear licence, but also non-nuclear sites where enhanced levels of radioactivity occur due to the presence of natural radiation sources. The identification of the nuclear sites is based on an inventory of the nuclear licenses as they have been granted by the competent authorities. As the non-nuclear sites do not require such license, their identification necessitates a specific methodology.

This presentation outlines the methodology developed by ONDRAF/NIRAS and SCK•CEN for identifying non-nuclear sites with enhanced levels of radioactivity.

The methodology combines data from an airborne radiometric survey of Belgium and

a literature search in order to identify and locate the non-nuclear sites. This proves



to be an extensive task undertaken by a taskforce combining the expertise of Controlatom (Belgium), NRG (the Netherlands), ONDRAF / NIRAS and SCK•CEN.



# **THE EFFECT OF EARTHQUAKE-INDUCED RADON RELEASE ON THE POPULATION IN THE SEISMIC ACTIVE REGIONS OF ARMENIA**

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For the first time on the basis of Spitak earthquake (Armenia, December 1988) experience it is found out that the earthquake causes intensive and prolonged radon splashes which, rapidly dispersing in the open space of close-to-earth atmosphere, are contrastingly displayed in covered premises (dwelling houses, schools, kindergartens) even if they are at considerable distance from the epicenter of the earthquake, and this multiplies the radiation influence on the population. The interval of splashes includes the period starting from the first fore-shock and ending by the last after-shock, i.e. several months. The area affected by radiation is larger than the territory of Armenia. The scales of this impact on population is 12 times higher than the number of people injured in Spitak, Leninakan and other settlements (toll of injured – 25 000 people, diseases caused by high levels of radiation in non-adapted population – over 300 000). The radiations of the influence are in direct correlation with the force of earthquake.

The basis for conclusion was the set of data on indoor radon monitoring in Yerevan City beginning from 1987 (120 km from epicenter) 5450 measurements and multivariate analysis with identification of cause-and-effect linkages between geodynamics of indoor radon under stable and unstable conditions of Earth crust, behavior of radon in different geological media during earthquakes, levels of room radon concentrations and effective equivalent dose of radiation impact of radiation dose on health and statistical information of the Ministry of Health on public health, etc.

As we have discovered, calculations of equivalent radiation dose show that compared with 1987, 1991 is character is by lower radon and its radio nuclides made up about 16mSv, i.e. 50 times more than mean radon equivalent radiation dose of the above-mentioned years. In the evidential part of the earthquake hearth the level of background radiation in dwelling houses as well as radon effective radiation dose radon, was many times higher. The interval of radon action is from first foreshock to the last aftershock, i.e. several months.

The following facts which have not received explanation up till now can be considered as consequences of long-lasting radiation influence on human organism: long-lasting state of apathy and indifference typical for the population of Armenia during the period of more than a year after the earthquake, prevalence of mali-



gnant cancer forms in disaster zones, prevalence of lungs cancer over the other forms of oncology diseases, etc.

The danger of the influence of natural radiation provoked by the earthquake exists for all urban territories of seism active regions, and it requires coordination of investigations in different countries according to precisely-elaborated programmers.



# **$^{210}\text{Po}$ AND $^{210}\text{Pb}$ IN ENVIRONMENTAL SAMPLES IN FINNISH LAPLAND**

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$^{210}\text{Po}$  and  $^{210}\text{Pb}$  are members of the  $^{238}\text{U}$  decay chain.  $^{210}\text{Po}$  ( $T_{1/2} = 138$  d) is formed by the decay of  $^{210}\text{Pb}$  ( $T_{1/2} = 22$  y) via  $^{210}\text{Bi}$  ( $T_{1/2} = 5$  d). In the atmosphere  $^{222}\text{Rn}$  forms its decay products  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  which are deposited on the ground and thus enter the human food-chain. In Finland most exposed are the Sami people having substantial intakes of reindeer meat.

In the Regional Laboratory in Northern Finland, measurements of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity concentrations in different environmental samples from Finnish Lapland have been performed. Especially the arctic lichen-reindeer-man food chain has been studied. The samples spiked with  $^{208}\text{Po}$ -tracer are leached by  $\text{HNO}_3$  and  $\text{HCl}$ . After the solution is made slightly acid, ascorbic acid and hydrazine is added and polonium is spontaneously deposited on nickel planchette. The  $^{210}\text{Po}$ -activity is measured by  $\alpha$ -spectrometry.  $^{210}\text{Pb}$  is determined from the same solution about 6 months later by carrying out another polonium deposition.

In this work plant and reindeer samples were collected early eighties and after Chernobyl accident in the Finnish reindeer herding area. The results of  $^{210}\text{Po}$  and



$^{210}\text{Pb}$  activity concentrations and the ratio of  $^{210}\text{Po} / ^{210}\text{Pb}$  in the plant and reindeer samples are presented.



# **NORM IN DRINKING WATER – MEASUREMENTS USING ULTRA LOW-LEVEL LIQUID SCINTILLATION SPECTROMETRY AND CONSIDERATIONS FOR COMPLIANCE WITH THE EU DRINKING WATER DIRECTIVE**

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The European Union Directive on the Quality of Drinking Water requests member states to control drinking water also for its radioactivity. The implications of the Directive on the radiological survey of drinking water in Austria will be discussed. NORM usually causes the by far largest contribution to the population dose and artificial radionuclides can in most circumstances be disregarded in drinking water. A simple, time saving and cheap method has been developed, which allows for simultaneous determination of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{210}\text{Pb}$  by using ultra low-level liquid scintillation spectrometry. Also tritium and  $^{222}\text{Rn}$  (if required) can be easily measured by LSC.

The Directive establishes a so-called "Indicative Dose": If the dose due to ingestion of drinking water might exceed 0.1 mSv/y it should be considered,





whether the dose should be reduced by countermeasures. The requirements for measurement methods and the strategy to determine compliance with the Indicative Dose will be discussed.



# RADIUM IN GROUND WATER CLOSE TO BUENA LAGOON IN COASTAL ZONE OF RIO DE JANEIRO STATE, BRAZIL

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Ground water close to the head of Buena Lagoon has high salinity and low pH values. Radium is present predominantly as  $^{228}\text{Ra}$ . Factors responsible for high radium mobility in ground water seem to be high salinity with resulting competition for adsorption sites, and low pH values with resulting limited adsorption of  $\text{Ra}^{2+}$  on positively charged surface of adsorbents. Behavior of uranium and thorium is also influenced by their speciation and low pH conditions. Uranium is present as positively charged uranyl ion  $\text{UO}_2^{2+}$  in low pH samples and is very mobile. Similarly, presence of positively charged  $\text{Th}^{4+}$  and thorium complexes with sulfate result in relatively high thorium concentrations at low pH range. On the other hand, mobility of phosphate released from dissolving monazite is probably reduced due to its adsorption and precipitation close to its source.



**Keywords** Groundwater, Coastal aquifer, Water geochemistry, Natural radioactive isotopes, Rio de Janeiro



# IMPROVING CRITERIA FOR REMEDIATION OF MONAZITE BY-PRODUCTS CONTAMINATED SITES IN BRAZIL

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In 1999 the first Brazilian decommissioning process occurred in São Paulo State at a monazite physical and chemical treatment plant, closed down since 1992. The decommissioning process comprised equipment, piping, buildings and land decontamination with the final site release for unrestricted use. At the present time, another monazite site is being decommissioned in the same State. Regulations for intervention situation in mining and milling of ores with uranium and thorium associated are being developed now, due to this, the decommissioning activities have been based on case-by-case analysis and ALARA criteria. Investigations are being made to propose suggestions for public participation in the process and establish a method of working together, in order to obtain a more effective communication between regulatory agency and society.

This paper presents an overview on the situation of three sites in São Paulo State: a past and a present remediation case and a site to be decontaminated in the future. All sites presented contamination with radioactive residues produced by out-



dated monazite chemical processing. We also propose some proceedings improvement based on past experience and international guidelines. This paper also suggest the implementation of public participation in the remediation process evaluation.



# ENVIRONMENTAL ASSESSMENT OF THE MATERIAL DEPOSITED ON THE FORMER URANIUM MINING DISPOSAL DUMP IN RADONIÓW

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Radoniów is a small town located in Lubomierz district near Jelenia Góra (south-west part of Poland). Beginning from the end of World War II up to the end of sixties there were uranium mining activities in the vicinity of the town. Uranium deposits were almost entirely exploited and the traces of the mining activities are the post-uranium dumps on the east side of mount Głębiec. The area of the terrain is about 6,85 ha. The material present on the dumps (containing high amount of uranium) is to be – according to the district authorities plans - used as the bedding for the construction of the road around the town (PHARE contract).

The measurements performed by the Central Laboratory for Radiological Protection (CLOR) and ordered by MOSTOSTAL-Warszawa (developer of the road) were to assess the usability of the dumped material for road construction.



The assessment program consisted of:  
environmental measurements at the disposal site (dumps) of the uranium mine in Radoniów;  
assessment of the usability of the materials from the dumping site for the construction of the circular road passing by Radoniów (part of the state road No. 30);  
analysis of the risk for the workers having contact with the material deposited on the dumps in Radoniów.

The following tasks were performed:

- measurement of the whole area by the mobile spectrometric laboratory (creating the radiological map of the area);
- measurements of the beta radioactivity at the surface of the material disposed on Radoniów dumps in 41 measurement points (RKP-1 radiometer);
- measurements of the gamma dose rate in 41 points (RKP-1 radiometer);
- measurements of the gamma dose rate in 41 points (Exploranium GR-130);
- measurements of the gamma dose rate in 5 points (ionization chamber);
- measurement of the gamma spectra in 3 points on the dumps and 2 points representing local background;
- sampling for further analysis in the laboratories of the CLOR (gamma spectrometry performed using natural radioactive contamination analyzers AZAR-90 and MAZAR-95).

The following samples were taken:

- 41 samples of the surface material of the dump (10 cm depth);
- 10 samples from the deep layers of the dump (taken by the specialized geological company "Geological Services");
- 2 samples representing local natural environment.

Every sample of the surface material of the dump was averaged from the 5 to 7 points collected from the central point and the points situated in the distance of 2 meters from the center.

Detailed description of the measurement methods, the results and their interpretation are presented in the poster.