Underground Measurements of Environmentala Radioactivity : European Examples

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Underground Measurements of Environmental Radioactivity: European Examples

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Abstract - Underground gamma-ray spectrometry is a fundamental tool for measurement of environmental radioactivity. The extremely low background in underground laboratories opens the possibility of performing studies which otherwise need more complex techniques, e.g. radiochemistry, albeit at the cost of long measurement times. This paper gives a brief overview of the current status of the technique in Europe and exemplifies its use by describing some recent studies of environmental radioactivity.

I. Introduction

Performing gamma-ray spectrometry underground significantly reduces the background. The background is due to radioactive materials in and around the detector, direct cosmic radiation and cosmogenic activation. It is the cosmic ray related background that is reduced by going underground. At great depth, the effect of cosmic rays is effectively eliminated and other sources dominate the background [1].

By lowering the background the sensitivity is improved and so this greatly enhances studies involving environmental radioactivity. The first underground studies using HPGe-detectors were in the field of fundamental nuclear physics and were related to detection of rare events such as the double beta decay or dark matter [2, 3, 4, 5]. These studies triggered developments in ultra low-background techniques and helped to identify impediments to further reductions in the gamma spectrometry background. Amongst the first underground measurements of environmental radioactivity were radiopurity studies aimed at selecting materials for large neutrino experiments [6]. The first underground gamma-ray spectrometry studies found that the limiting factor is often radioactivity in the HPGe-detector itself [7, 8, 9, 10]. In recent years, internal radioactivity in state-of-the-art commercial detectors has been considerably reduced and consequently laboratories with programs on ULGS (ultra low-level gamma-ray spectrometry) [11] are now found in several places in Europe. A network of several of these laboratories called CELLAR (Collaboration of European Low-level underground LAboRatories) has been created in order to promote underground radioactivity studies. It is a natural step in low-level radioactivity measurements to work in networks for the sake of sharing resources as measurement times are long and studies with any urgency will generally exceed the capacity of any one laboratory. An overload of measurements at one laboratory can thus be overcome. The CELLAR laboratories have different primary responsibilities, which is a sign that the field of underground studies is extending its usefulness from the field of fundamental physics to many other disciplines including environmental radioactivity studies and reference measurements. Table I shows that only three laboratories, LSCE, VKTA and IAEA-MEL have environmental radioactivity as their primary role, see e.g. [12, 13, 14], but other laboratories undertakes reference measurements which frequently are on environmental samples. This paper aims at giving an overview of the current situation in Europe and describing some recent applications in the field of environmental radioactivity.

II. Materials and methods

A. HPGe-detector systems

In all the CELLAR laboratories a typical set-up for an HPGe-detector is:

- Ultra low-background HPGe (often of large volume).
- 15-25 lead shield of which the inner 2-5 cm is low in ^{210}Pb (< 5 Bq/kg)
- Inner lining of 1-15 cm of freshly produced electrolytic copper. If the radioactivity of the lead is very low the copper shield need not be very thick.

Such a detector system is not suitable for above ground placement. Muons and secondary neutrons will increase the background if the shield is too thick and the Cu is better left out completely if the lead quality is very high. The CELLAR laboratories located at shallow depth use anti-muon shields of plastic scintillators or gas proportional counters in order to reduce the background from cosmic rays [15].

Detector windows are mostly ultrapure Al (KryAl) or electrolytic Cu. Other radiopure materials have also been tried, e.g. Si. Recently commercial detectors have become available with carbon epoxy windows. These windows have proven mechanically strong while causing very little attenuation to low energy gamma-rays and X-rays. It is, however, difficult to eliminate ^{40}K in carbon epoxy windows. This inhibits the use of these windows in very deep laboratories [16].

TABLE I
List of the underground facility of the CELLAR network members.

Institute	Underground laboratory	Depth (m w.e.)	Stratum	Country	Main activity
LSCE [12]	Modane	4800	Rock	France	Environmental radioactivity
LNGS [17]	Gran Sasso	3800	Rock	Italy	Radiopurity of construction materials to support to rare event experiments
PTB [18]	UDO in the salt mine Asse	2100	Salt	Germany	Reference measurements
IRMM [19]	HADES	500	Clay	Belgium	Reference measurements
University of Iceland		350	Water, sediment and rock	Iceland	Studies of background components in radiation detectors
VKTA [20]	Felsenkeller	110	Rock	Germany	Environmental radioactivity
IAEA-MEL [21]	CAVE	30	Rock and concrete	Monaco	Environmental radioactivity
Max Planck Institut für Kernphysik, Heidelberg [22]	Low-level laboratory (and Gran Sasso)	15 (3800)	Rock soil and concrete (Rock)	Germany (Italy)	Rare events research and detector development

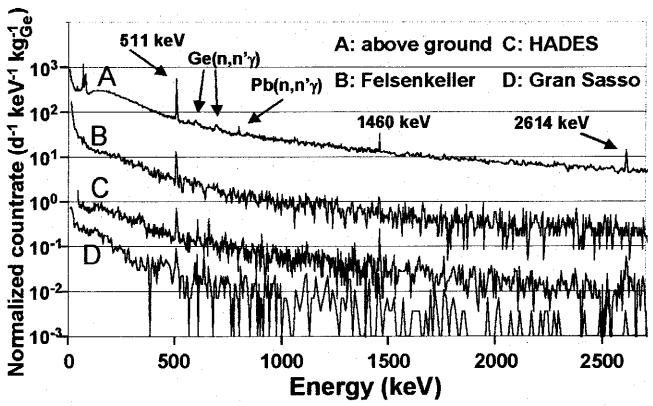

Fig. 1. The background countrate normalised to the Ge-crystal mass for different detectors used for studies of environmental radioactivity [19, 20,23]

B. *Background comparison*

Figure 1 shows the background spectra in 4 different underground laboratories (using 4 different detectors with only passive shielding). The spectra are normalised using Ge-crystal mass. As all detectors are of similar shape (coaxial), it is a reasonable approach to use the Ge-crystal mass to allow comparison. It is evident in Fig. 1 that the the cosmic ray induced continuum is significantly reduced at greater depths. In a good low-background system above ground or at shallow depth it should be difficult to see peaks other than the annihilation peak at 511 keV and the peaks from neutron capture and scattered neutrons at e.g. 596 and 691 keV [24]. At deep locations, the purity of the detector and shield becomes more important, provided that the radon-gas is efficiently expelled from the counting volume. Figure 2 shows examples of the integrated background countrate from 40 to 2700 keV for some HPGe-detectors used in CELLAR laboratories for the measurement of environmental radioactivity. It is evident from Figure 2 that at very deep locations the level of background is determined by the radiopurity of the detector and the activation and spallation products remaining from the above ground storage/production. Except for gamma-rays, X-rays and Bremsstrahlung arising from events some distance away from the Ge-crystal, the spectrum may contain counts from direct detection of alpha or beta particles from impurities in the Ge-crystal, detector window or cryostat. Examples of such events are beta particles from ^{40}K (max energy 1312 keV), ^{210}Bi (daughter of ^{210}Pb with □⁻-max energy of 1161 keV) and ^{60}Co (max-energy 2824 keV). In the best systems the background is set by ^{68}Ge, in which the positron from the decay of the daughter ^{68}Ga deposits its energy in the Ge-crystal. In a special case (Ge enriched in ^{76}Ge) a rare event, such as the two neutrino double beta decay of ^{76}Ge, forms a significant part of the background [4, 5], which amounts to less than 10 counts per day per kg Ge (40-2700 keV).

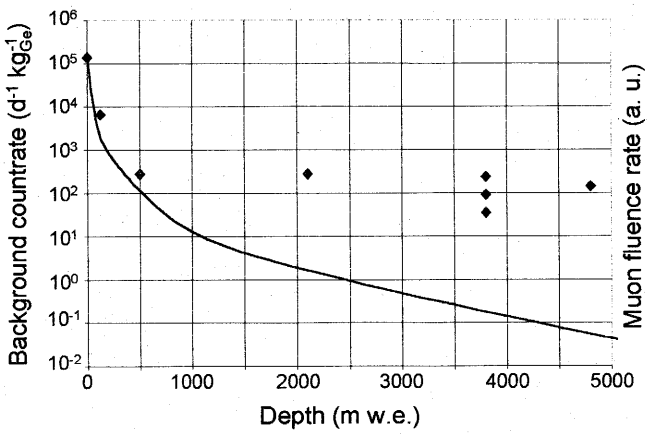

Fig. 2. The integrated background countrate from 40 to 2700 keV for HPGe detectors in some CELLAR laboratories. The solid line shows the normalised muon fluence rate in arbitrary units.

With a good design of the passive shield and the use of an anti-muon shield it is possible to achieve an integrated background countrate in the order of 10^4 d^{-1} kg^{-1}(Ge) above ground [25, 26]. This is comparable to a detector equipped with only a passive shield at a depth of some 50-100 m w.e. Although an active shield can effectively reduce the direct impacts of the muons, it is not possible to alleviate the cosmogenic activation. Radionuclides like ^{68}Ge, ^{65}Zn, ^{57}Co and ^{54}Mn will be produced in the Ge-crystal and ^{59}Fe, ^{56}Co, ^{58}Co, ^{60}Co and ^{54}Mn in the Cu. Other factors limiting the effectiveness of an active shield for background reduction are pulses from delayed neutrons and gaps in the veto shield. As the veto detector also adds to the complexity of the system, there is a greater risk of technical problems and it may cause a need for accidental coincidence corrections. The ease of making measurements is an essential factor to consider when making frequent accurate measurements of environmental radioactivity at a moderate cost.

III. Studies of Environmental Radioactivity

A. Radioactivity in human bones

It was recently shown that by using ULGS in the underground laboratory HADES (-500 m w.e.) it is possible to measure natural radioactivity from bones of "non-exposed" persons *in-vitro*. Quantification was possible even when only small samples (1-10 g) were available [27, 28]. This information is essential for definition of the baseline of radioactivity in human bones of "non-exposed " individuals. In bones of highly exposed persons, where the activity is easier to assess when measured *in-vitro*, the excessive uptake is usually from one specific source instead of a combination of sources. It is thus not always justified to extrapolate from highly exposed individuals to those with only exposure to normal sources. Of main interest for this study was ^{210}Pb, which is a bone seeking daughter in the decay chain of ^{222}Rn. Several investigations on *in-vivo* retrospective assessment of ^{222}Rn-exposure have been carried out world-wide. It is unclear which bone mass is most suited for such studies, the skull or the knee. The distribution of ^{210}Pb in the bone is also not well known. Many models assume a homogeneous volume distribution although recent results show that a homogeneous distribution within the bone mineral (hydroxyapatite) is more realistic [28]. This approach is important since the 46.5 keV gamma-rays from the decay of ^{210}Pb are heavily attenuated in the bone. The bone itself is an inhomogeneous structure. In a simplified model it can be divided in two layers. The outer layer (cortical bone) holds most of the mass and forms the supporting structure. The inner part of the bone (trabecular bone) is spungiform and hollow in order to make room for blood vessels and soft tissue.

One of the advantages of gamma-ray spectrometry using HPGe-detectors, is that a measurement generates information on all gamma-ray emitting radionuclides (whether detected or not). The recent study at IRMM focussed on ^{210}Pb but could also generate data for Th-daughters (^{228}Ra and ^{228}Th), ^{40}K, ^{137}Cs and ^{226}Ra (via the ^{222}Rn-daughters).

A more direct way of measuring Rn-exposure retrospectively is to measure ^{210}Pb in lung cells. In Germany there are many thousands of lung samples (freeze dried, homogenised and sieved at <0.5 mm) from former uranium-miners. It is possible to generate valuable data that may be useful for assessing the impact of ^{222}Rn on the human body by measuring ^{210}Pb in these samples. A problem is that the samples' masses are small, in the order of 1-3 g. It was shown in a feasibility study that the ^{210}Pb concentration for a few samples were in the order of 1-20 mBq/g. This could be measured in HADES at 500 m w.e. but was not detectable in Felsenkeller at 110 m w.e. The detection limit in HADES for a typical sample of 1.4 g was 1 mBq/g after a 4 week measurement.

B. Environmental neutron fluence

The foil activation method has been used for many years to measure neutron fluence rates up 10^{15} $s^{-1}\cdot cm^{-2}$ in nuclear reactors. By using thicker foils (discs) and activating them until saturation activity is reached and measuring them for a long time period on an HPGe-detector in an underground laboratory, it is possible to measure thermal neutron fluence rates down to 10^{-4} $s^{-1}\cdot cm^{-2}$ [29, 30, 31]. This is about a factor of 100 below the normal environmental level. Different metals can act as the targets. It is also possible to generate information on the neutron spectrum. At IRMM, this technique is referred to as the LAM (Low Activation Method) [32]. It has the potential of opening up for some interesting applications. In the field of radiation protection, metal discs can be used as simple low cost detectors that cannot fail. There is no need for electronics to control them and they are not sensitive to other types of radiation. They are small and can be placed in remote or difficult places. One can use such metal discs as backup or benchmarking for existing more complex neutron monitoring systems. The technique is used at IRMM and SCK•CEN (the Belgian nuclear centre in Mol) to better quantify the neutron fluence outside artificial neutron sources to assure doses are extremely low in neighbouring communities even though the neutron fluence is known to be well below the legislated limit [30, 33]. Other frequently found sites that are important to monitor are cyclotrons and linacs for use in hospitals [34].

During the JCO accident in 1999, an uncontrolled neutron field irradiated the surroundings for about 20 hours. After the accident the independent investigation team collected samples from the surroundings in order to retrospectively assess the neutron fluence [35, 36]. Some of the samples had low activity because of their distance from the criticality site or due to a low neutron capture cross section. Certain samples required low-level gamma-ray spectrometry. These time-consuming low-level measurements greatly restrict throughput. In order to be able to measure ^{51}Cr (half-life: 28 days) in steel spoons [37] and ^{140}Ba (half-life: 12.8 days) via its daughter ^{140}La, in air-filter samples, the independent team

collaborated with IRMM and VKTA. Legislation in most countries states that in case of significant radiological incident e.g. a criticality accident, the authorities need to investigate the causes and the impact of the accident. Having small metal discs distributed around nuclear facilities may simplify such tasks as well as providing accurate feedback on neutron fluences during normal operation.

In isotope geology it is essential to have detailed knowledge of cosmogenic production rates [38]. These rates vary with the depth in matter [39]. Using thin discs it is possible to study environmental cosmogenic production inside various matrices. Although induced disc activities may be sufficiently high to allow measurement in a good detector above ground it is essential to measure underground to stop the target from being further activated after its removal from the site being monitored. The results of future investigations in this field may prove valuable also for work in designing shields for detectors measuring low-levels of radioactivity.

C. *Underground studies for benchmarking purposes*

IRMM has performed ULGS since 1992. Initial studies were directed towards a better understanding of background components. Measurements of environmental radioactivity have since formed a major part of the work program [40, 41]. Since 1999, interest has been directed towards performing reference measurements to benchmark other methods such as those based on radiochemistry or mass spectrometry. Any technique can produce an unwanted bias in certain cases. It is not always known that this is the case and it might never be revealed unless another independent technique is employed for benchmarking. Techniques based on radiochemistry and mass spectrometry often require extensive and complicated sample preparation. It is often necessary to make assumptions that appear obvious but are difficult to verify objectively. By using ULGS it is possible to undertake gamma-ray spectrometry on samples that require extremely simple or even no sample preparation. Long-lived radionuclides are usually best measured using mass spectrometry, whilst short-lived ones are better measured using radiometric techniques. The range of half lives between these two zones is quite wide and depends on many factors, but stretches approximately from some hundred years to some ten thousand years. With ULGS the usefulness of gamma-ray spectrometry extends to cover also low activities of very long-lived radionuclides such as ^{26}Al (half-life: 710,000 a) and can in certain cases compete with other techniques.

Some recent examples of where ULGS benchmarking exercises of other methods were called upon are:

(i) Accelerator Mass Spectrometry (AMS) measurements of ^{26}Al in meteorite samples [42, 43].

(ii) Gamma-ray spectrometry preceded by radiochemical separation [44, 45] to measure ^{60}Co produced in steel at Hiroshima during the A-bomb in 1945.

(iii) Measurement of the ^{94}Mo(n,p)^{94}Nb nuclear reaction cross section [46] using radiochemical separation of Nb from the Mo target.

(iv) Resolving discrepancies between Electron Raman Scattering and Glow Discharge Mass Spectrometry [47].

D. *^{226}Ra and ^{228}Ra in water for human consumption*

Modern radiation protection limitations require the simultaneous and routine determination of ^{226}Ra and ^{228}Ra in water for human consumption. Although the regulations for various radionuclides in different countries are not alike, they are based on a maximum ingestion dose (effective dose) of typically 0.1 mSv a^{-1}. Table II shows activity concentrations for intake of ^{226}Ra and ^{228}Ra, which will result in reaching this dose limit. A common convention is that the analytical sensitivity is at least a factor of 5-10 lower than the limit. For this reason, the requirements for detection limits for the analysis of ^{226}Ra and ^{228}Ra were set to 10 mBq L^{-1} and 2 mBq L^{-1}, respectively.

Instead of α-counting in Lucas cells for ^{226}Ra and additional β-counting for ^{228}Ra, a simultaneous method to determine both radium nuclides by low-level γ-ray spectrometry after radiochemical separation [48] was applied. ^{226}Ra and ^{228}Ra were co-precipitated with Ba/Pb and subsequently filtered. The precipitate was sealed in a radon-tight quartz ampoule. After secular equilibrium was reached, ^{226}Ra was determined using the γ-transitions of the daughter nuclides ^{214}Pb and ^{214}Bi. ^{228}Ra was quantified from analysis of the daughter ^{228}Ac (Table III).

By counting a sample of 1 L for 1 day using a well-type HPGe-detector in the underground laboratory Felsenkeller, detection limits of 7.4 mBq L^{-1} for ^{226}Ra and 2 mBq L^{-1} for ^{228}Ra were reached.

In a similar study on drinking water, Semkow [49] points out that using underground HPGe-detectors for the measurement of ^{228}Ra, will considerably improve the measurement throughput.

TABLE II
Activity concentrations in water (mBq L^{-1}) that would generate an ingestion dose of 0.1 mSv a^{-1}.

Radio-nuclide	Germany [50]		European Union [51]
	< 1 a child	adult	-
^{226}Ra	125	1000	500
^{228}Ra	20	410	200

TABLE III
γ-energies used for ^{226}Ra and ^{228}Ra determination

Nuclide	Measured Radionuclides	Half-life	E_γ (keV)
^{226}Ra	^{214}Pb, ^{214}Bi	1600 a	295, 351 609, 1120, 1764
^{228}Ra	^{228}Ac	5.8 a	338, 911, 968

IV. Summary and Discussion

Great advances in ultra low-level measurements techniques using HPGe-detectors were made in the late 1980s and the 1990s. The background of the best HPGe detector systems may be reduced even further in very deep laboratories (deeper than 1000 m w.e.) by using very expensive germanium isotopically enriched in e.g. ^{72,73,74}Ge. For environmental studies, rarely will there be great value from achieving these very low background count rates as measurement times of months are required to benefit from such a reduction in background. In many environmental studies, measurement times longer than a week are not practical or even feasible.

Some of the large and deep laboratories have plans to extend their laboratories. For 'big science' experiments studying rare events, gamma-ray spectrometry measurements have become an essential part of the infrastructure needed to set up a large experiment.

At several places, shallow (10-30 m w.e.) laboratories with easy access are being set-up. The authors perceive a trend that in these laboratories, as well as at deeper sites, HPGe-detectors will become the working horses for environmental studies. Using high efficiency detectors and coincidence techniques (anti-Compton, anti muon) will generate valuable data at a higher rate and often more reliably than using above ground systems or more complex techniques based on radiochemistry or mass spectrometry. As a result it should for example be possible to give faster responses in emergency situations as well as to better enforce legislation on environmental issues.

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