MetroERM radiological early warning

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TABLE OF CONTENTS

1 Executive Summary

Introduction

All European countries operate radiological early warning networks, and there are approximately 5500 dosimetry monitoring stations currently active across Europe. However, many of these stations are based on simple detector designs which do not give the required level of radiological accuracy or detail, and thus further time-consuming data analysis is needed before any decisive action can be taken. To address this, this project developed devices, new measurement techniques and methods to coordinate the radiological data collected from monitoring stations and to improve comparisons between different stations and networks. This will result in the faster and more coordinated response of European authorities in the event of a nuclear emergency.

The Problem

During an airborne spread of nuclear contamination there is an urgent need for authorities to advise the population on the necessary precautions to stay safe. Recommendations from the European authorities could affect millions of people and may have severe economic and sociological consequences. Therefore, metrologically sound monitoring data of ambient dose rate and airborne radionuclide activity concentrations are a prerequisite for sound governmental decisions.

Safety standards for the health protection of the general public and workers against the dangers arising from ionising radiation are laid down in the Council Directive 2013/59/EURATOM and are mandatory for all EU Member States. In addition, as a direct consequence of the Chernobyl accident, information exchange in the event of a radiological emergency is implemented by the European Community Urgent Radiological Information Exchange System (ECURIE) operated by the European Commission. The 5500 dose rate monitoring stations operating in Europe provide hourly data transmission to the European Radiological Data Exchange Platform (EURDEP). In the case of a major radiological or nuclear accident, the information collected by EURDEP is used by the ECURIE system to initiate the responses of national authorities to radioactive contamination.

Networked radioactive contamination monitoring stations use two main types of detectors to determine if levels of airborne contamination are increasing as a result of a nuclear incident:

- Dose stations use passive devices to monitor for changes in the levels of radioactivity in the environment. Current dose meters are not capable of providing data on specific radionuclides, but recent developments in detector materials mean that modern instruments have the potential to provide this information.
- Airborne particulate samplers use a pump operating at high flow rates to suck air through a filter and trap airborne particles and often augment dose monitoring. After many hours of operation filters are removed for offsite analysis using chemical extraction techniques. The analysis methods used provide very accurate determinations of radioactive contamination but there is a significant delay in generating contamination level results.

All monitoring systems are affected by background effects such as radon, a naturally occurring radioactive gas, and cosmic interferences. New detector materials are becoming available but these need to be performance evaluated for dose and determination of radionuclides before they can be used in instruments and devices for active radioactive monitoring. The most accurate gamma radiation detectors HPGe (High Purity Germanium) have previously been restricted to laboratory conditions because they need to be cooled to liquid nitrogen temperatures. However, new refrigeration techniques can now be used which means the HPGe detectors can be deployed at monitoring stations.

Evaluation of these technologies is needed to provide cost effective recommendations for monitoring networks. Prior to this project, it was common to see uncertainties in radiological data of a factor of 2 or more. Therefore, automation, an upgrade to newer detector types, accurate characterisation and standardised operation procedures to ensure trans-boundary events are accurately recorded across the EU are needed in order to ensure that the best possible data is available to ECURIE.

The Solution

The aim of this project was to improve the metrological foundation of measurements (devices and methods) for monitoring airborne radioactivity and to introduce pan-European harmonisation in data reliability for area dose rate measurements which are input to EURDEP and other monitoring networks. In addition, novel and improved instrumentation and new measurement techniques and analysis methods were developed for field stations of the radiological early warning networks in Europe. The characteristics and properties of newly developed spectro-dosemeters were investigated and published. These publications include recommendations how the novel instruments should be integrated in the existing infrastructure. The results achieved clearly demonstrate the ability of the novel spectrometric detectors to replace conventional dose rate meters like Geiger Muller based dose rate meters presently mostly used in early warning networks. Such a modernization would be accompanied by a considerable increase of the accuracy of the data and its information content (provision of nuclide specific data). Similar to dose rate monitoring, novel spectrometric detectors with medium energy resolution like $LaBr_3$, $CeBr_3$ and SrI_2 have been investigated experimentally as well as by various Monte Carlo simulations for their capability to analyse activity concentrations of radioactive particulates collected at the filters of air samplers. Three novel air samplers (two compact portable and one modular transportable) have been developed and tested. All are on-line capable and provide data in real-time.

Impact

The results obtained in the project represent important steps towards the harmonization of radiological data provided by the early warning network stations in Europe and by environmental radiation monitoring, in general. Specifically:

- More precise area dose rate and air borne radioactivity monitoring data can be derived
- New measurement techniques and traceable measurement procedures are available
- Basic metrological information (performance parameters) is provided for dosimetry, spectrometry and air sampler systems for metrologically sound measurements of dose rates, contamination levels on ground and air contamination levels, in real-time
- Metrological developments and improvements allow an early indication of a nuclear or radiological threat
- Best practice derived from comparison exercises and evaluation of differing calibration procedures is disseminated via e-learning material made permanently available on the internet for the user communities.

In addition, the metrological infrastructure in Europe for dosimetry at low dose rates, has been improved by the establishment of a second underground low dose rate calibration facility, operated by IFIN-HH in Romania.

2 Project context, rationale and objectives

2.1 Context

The nuclear power plant accident in Chernobyl (Ukraine) in 1986, with massive transboundary implications, led to severe medical, environmental, economic and sociological consequences for the affected countries. The huge amount of radioactivity released after the accident, combined with meteorological factors, caused large-scale contaminations not only in Ukraine and its neighbouring countries Russia and Belarus, but also in Scandinavia and less severe in most other European countries. Parts of the large contaminated areas (e.g. 280.000 km^2 in Belarus) had to be, at least temporarily, excluded from agricultural production. Therefore, the economic costs of this accident were tremendous. Estimates are in the order of more than 100 billion dollars for Ukraine and 35 billion dollars for Belarus. Moreover, as pointed out by the World Health Organisation (WHO): "*In addition to the lack of reliable information provided to people affected in the first few years after the accident, there was widespread mistrust of official information and the false attribution of most health problems to radiation exposure from Chernobyl*".

As a direct consequence of the Chernobyl nuclear power plant accident, the Council of the European Union has adopted the Council Decision 87/600/Euratom which lays down basic requirements for "…the early exchange of information in the event of a radiological emergency" (European Commission 1987). Since 1987, all European countries have installed automatic dosimetry network stations as well as air sampling systems for the monitoring of airborne radioactivity. In Europe, at present, about 5500 stations measure dose rate

values in nearly real time. In addition, a few hundred air samplers are operated. Most of them need extended accumulation times with no real-time measurement capability. National dose rate data are provided on an hourly basis to EC via EURDEP, and a web interface with reduced functionality supplies information to the general public.

In case of a nuclear emergency with transboundary implications, the EC may issue important recommendations to EU member states based on the radiological data collected by EURDEP. These recommendations may affect millions of people and could have severe economic and sociological consequences. Therefore, the reliability of the EURDEP data is of key importance. Unfortunately, the dose rate and activity concentration data are neither validated nor harmonized between the different networks.

On the other hand, reliable radiological data from routine measurements are also a prerequisite in order to achieve credibility and acceptance by the public concerning the operation of nuclear power stations. Unfortunately, almost three decades after the Chernobyl accident, the non-validated radiological data provided by EURDEP still show significant (artificial) differences at the borders of neighbouring countries, related to different instrument properties and measuring procedures. For these reasons, the European Commission Joint Research Centre (JRC) in Ispra (Italy) and the European Radiation Dosimetry Group (EURADOS) have undertaken considerable efforts in the last decade to harmonize the data of dosimetric network stations in Europe.

In 2011, the nuclear power plant accidents in Fukushima once more demonstrated the indispensable need for reliable environmental radiation monitoring. Although this remote accident did not lead to any measurable increase in the ambient dose equivalent rates in Europe, a pronounced increase in the radioactivity concentrations in air, especially of ^{131}I, ^{137}Cs and ^{134}Cs, was observed on the filters of high-volume air samplers. Comparisons of the reported values showed inconsistencies and again revealed the need for harmonization of reported results. In the event of a major radiological emergency, the early and reliable knowledge of radioactivity concentrations in air, and subsequently the assessment of contamination levels of farmland and of dose rate levels in urban areas are of key importance for organising sound countermeasures for the protection of the general public from the dangers arising both from direct external radiation and from intake of radioactivity from ingestion or inhalation of contaminated food or air.

2.2 Objectives

This project set up to improve the metrological foundation of measurements (devices and methods) for monitoring airborne radioactivity and to introduce pan-European harmonisation in data reliability for area dose rate measurements which are input to the European Radiological Data Exchange Platform (EURDEP) and other monitoring networks. This project addressed the following scientific and technical objectives:

1. To develop novel and improved dosimetry systems for field station use to enable both the measurement of dose rates and the collection of nuclide-specific information; including comprehensive scientific investigations of detector features and of spectra evaluation and deconvolution methods for new and improved measurement systems based on novel spectrometric detectors, e.g. $LaBr_3$, $CeBr_3$, SrI_2 and CdZnTe;
2. To validate the new techniques for the calculation of dose rates and contamination levels from in-situ gamma spectra by Monte Carlo simulations and bench mark experiments;
3. To undertake enhanced on-site evaluation of the diverse environmental and radiological conditions and measurement techniques used at dosimetry monitoring stations (provision of background information on site conditions and scientific development of appropriate correction methods);
4. To develop improved detection methods and data analyses techniques to enable accurate measurements of low activity concentrations of radon (in the range from 300 Bq m^{-3} and below). To develop and cross-check procedures for determining the blank indication of active radon monitors;
5. To develop novel and improved instrumentation for the field of airborne radioactive particulate monitoring, focused on mobile systems with real-time capability. This includes the development of novel traceable reference materials and standard sources (especially for large-area aerosol filters) and to perform proficiency tests and other comparison exercises to quantify airborne radioactivity measurements at field stations;

6. To validate the new techniques for the field of airborne radioactive particulate monitoring
7. To validate common metrological procedures and to implement traceable calibrations of detector systems used to supply data to central databases, especially EURDEP;
8. To develop new and more sophisticated data analysis protocols to enable rapid information dissemination;
9. To install an underground low-dose (≤100 nSv h^{-1}) calibration facility of IFIN-HH at Slanic-Prahova in Romania and to validate it against PTB's globally unique UDO II underground facility.

3 Research results

In the following, the results of this joint research project will be summarized and presented against each of the project's objectives.

In order to identify the current state of the art at the start of the project, different questionnaires have been developed and sent out to the operators of European early warning networks. The questions focused on the methods used to:

a) Detect airborne contaminations by monitors and samplers of radioactivity in air,

b) Monitor area dose rates,

c) Handle, transfer and store radiological data in real time and

d) Provide traceable calibrations of detector systems.

In addition, information on the needs of participants interested in an intercomparison between organizations involved in the detection of radioactive contaminations on a filter medium was collected. The answers received from these questionnaires have been carefully evaluated and provided important input for other investigations.

3.1 Novel and improved dosimetry systems for field station use, based on novel spectrometric detectors

To detect radiological incidents, all members of the European Union have installed nationwide radiological early warning networks. Most of the installed detector systems supply only dosimetric information. Novel spectrometry systems are considered to be good candidates for a new detector generation for environmental radiation monitoring because they will supply both nuclide-specific information and ambient dose equivalent rate values at once.

Therefore, within this project, novel and improved instrumentation and new measurement techniques and analysis methods were developed for field stations of the radiological early warning networks in Europe.

New spectrometry systems, based on scintillators like $LaBr_3$, $CeBr_3$, SrI_2, and the semiconductor CdZnTe, were characterised experimentally, as well as by various Monte Carlo (MC) simulations, so that they can function as dosemeters as well. The aim of using these spectro-dosemeters is to derive nuclide specific information additionally to the determination of area dose rates. The performance of the novel spectro-dosimetry systems has been tested under metrologically well-defined irradiation conditions, like e.g. in PTB's underground calibration facility UDO II and in quasi isotropic irradiation fields in radon-progeny atmospheres at PTB's former radon chamber. In addition, long term measurements of several months, under real weather conditions, were performed to study the feasibility to replace conventional dose rate meters by spectro-dosimetry systems. The measurement results were compared with data derived from well characterised reference instruments. Examples of some studies are shown in the subsections below.

In several publications, the characteristics and properties of spectro-dosemeters developed in this project were described. These documents also include recommendations for the integration of the novel instruments in the existing infrastructure of early warning networks. The basic properties of the detectors, like e.g. their inherent background, energy resolution and sensitivity were studied and the different detectors were compared with each other. Results which were achieved in one of these studies are listed in Table 1 [2]:

Table 1: Properties of detectors investigated and characterized at PTB.

Material of detector crystal	$LaBr_3$	$CeBr_3$	SrI_2	CdZnTe
Dimensions (cylindrical with diameter *d* and height *h*)	*d* = 2.54 cm *h* = 2.54 cm	*d* = 2.54 cm *h* = 2.54 cm	*d* = 2.54 cm *h* = 2.54 cm	cubic, 1 x 1 x 1 cm^3
Density/ (g cm^{-3})	5.18	5.07	4.6	6.34
Typical total light yield of photons / MeV^{-1}	66,000	43,000	85,000	-
Energy resolution at 662 keV/ %	2.7	4.5	4.1	2.6
Radioactive contamination	^{138}La and ^{227}Ac	^{227}Ac	none	none
Inherent background/ 10^3 h^{-1}	34.7	2.60	0.60	0.052
Inherent background / nSv h^{-1}	82	9.8	< 1	< 1

The results achieved clearly demonstrate the ability of the novel spectrometric detectors to replace conventional dose rate meters like conventional Geiger Muller based dose rate meters presently mostly used in early warning networks. Such a modernization would be accompanied by a considerable increase of the accuracy of the data and its information content (nuclide specific data).

In a different study, two spectrometric monitors, the LaBr3 (Ce)(1.5" x 1.5") based SpectroTRACER from Saphymo and the CdZnTe (1 cm^3) based GR1-A from Kromek (with the housing from Saphymo and assembled by BfS) were irradiated at different gamma fluence rates, energies and angles at UPC's secondary standard calibration laboratory using ^{241}Am, ^{137}Cs and ^{60}Co sources to study their linearity, energy resolution and angular response The detectors show almost an isotropic response from 90º (incident photon from the top) to ~ -30º (330º), while after this breakpoint the efficiency drops to nearly zero at -90º (270º). The reason for this is the absorption produced by the electronics of the monitor, located in the bottom half of the probes. Both spectrometric detectors work properly up to ambient dose equivalent rates of approximately 2 mSv h^{-1}.

3.1.1 Detector's inherent background

The inherent background of the spectro-dosimetric detectors was measured in UDO II, the low dose rate underground laboratory of PTB, which shows an ambient dose equivalent rate of 1.4 nSv/h only [2]. A lead castle, installed at UDO II, reduces the natural background radiation even further by a factor of about 10. Therefore, the inherent background spectra of the detectors were measured inside this lead castle.

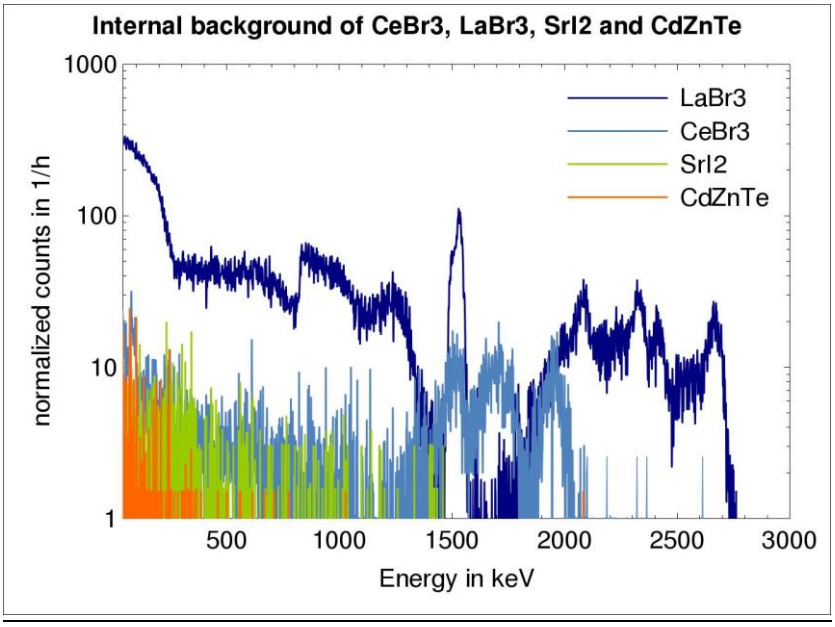

Figure 1: Inherent background of the different detector types measured in a lead castle in PTB's underground laboratory UDO II [2].

Background spectra of the four different detector systems described in Table 1, in the energy range from 30 keV to 3 MeV, are depicted in Figure 1. The upper energy limit was chosen to cover the highest occurring energy of the natural radiation of 2614 keV (^{208}Tl). The lower energy limit was set to 30 keV due to the absorption of the housing at lower energies and to cut off noise. Using the conversion coefficient from count rate to dose rate of Figure 3, the background spectra correspond to inherent background dose rate values of 82 nSv/h ($LaBr_3$), 10 nSv/h ($CeBr_3$) and less than 1 nSh/h for SrI_2 and CdZnTe, respectively.

3.1.2 Energy dependence of the response

The response of the various detectors with respect to different incident photon energies (60 keV (^{241}Am), 122 keV, 136 keV (^{57}Co), 662 keV (^{137}Cs), 1173 keV, 1332 keV (^{60}Co)) was measured in radiation fields at UDO II [2]. This sensitivity is a limiting factor for the time required for a reliable determination of activity concentrations and ambient dose rate values. The aim is to measure these in less than an hour. The four different photon fields cover the wide range of energies observed in the natural radiation. For a quantitative analysis the net count rate of the photo peaks was derived.

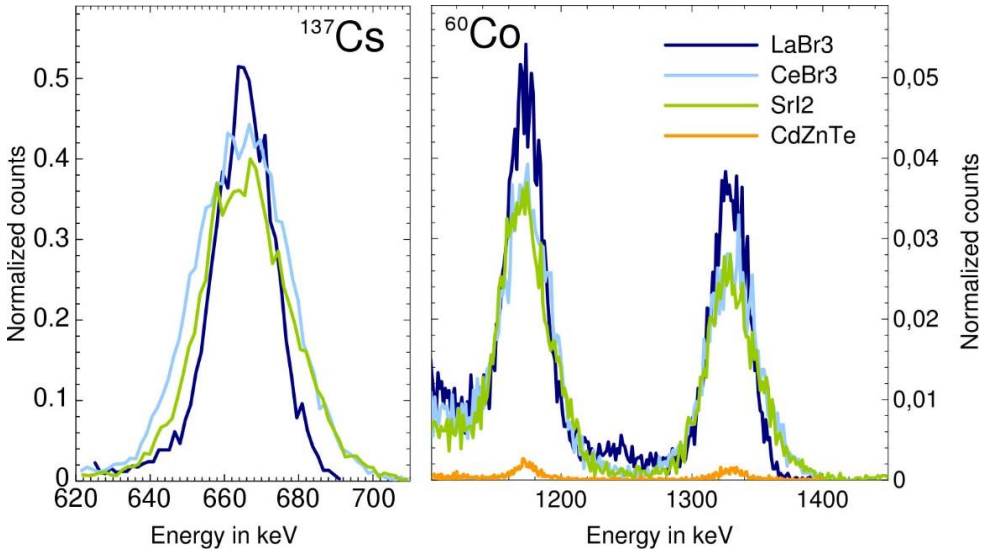

Figure 2: Response of the different detectors [2].

In Figure 2, the photo peaks found in the spectra of the four detector systems, described in Table 1, produced by ^{137}Cs (left) and ^{60}Co (right) are shown. The scintillators show a similar dependence of the net peak count rate on the photon fluence with differences of less than 10 %. The SrI_2 is less sensitive than the bromide detectors which is due to the lower density, as the geometry of the scintillation crystals is identical. The spectrum of the CdZnTe detector shows less than 10 % of the peak count rate at energies above 662 keV which is mainly caused by the smaller detector size. The exceptionally good energy resolution of 2.7 % at the 662 keV (^{137}Cs) peak of the $LaBr_3$ detector can be seen on the left in Figure 2, which is comparable to that of the CdZnTe detector, 2.6 %. The measured energy resolution of the $CeBr_3$ detector, 4.5 %, and of the SrI_2 detector, 4.1 %, are of the same order. However, a much better resolution for SrI_2 was reported in literature. The rather poor energy resolution of this measurement is most likely caused by the readout electronics, which was not optimized for processing the signals of a SrI_2 scintillator.

In Table 2, the dependence of the count rate per dose on the incident photon energy is quantified and summarized. The differences are observed both at the 60 keV peak of ^{241}Am and at the 122 keV peak of ^{57}Co, which are caused by the different materials and geometries of the detector housings. This means that in case of the $CeBr_3$ based detector a ^{137}Cs source (662 keV) that causes a dose rate of 5 nSv/h in the position of the detector has to be measured at least for 1 h to achieve a statistical uncertainty of 5 % of the peak area, inherent background effects not included.

Table 2: Measured counts of the peak integral per unit $H^*(10)$ of each detector at different energies (the standard uncertainties are listed with the expansion factor k = 1). The normalized photo peak efficiency can be calculated from the $H^*(10)$ values [2].

Peak Energy	Peak integral of detector based on:			
	$LaBr_3$	$CeBr_3$	SrI_2	CdZnTe
keV		(10^{11} Sv^{-1})		
60	59.1(7)	57.2(7)	46.6(5)	9.9(2)
122	43.6(6)	41.3(7)	34.4(4)	6.06(14)
662	2.27(3)	2.19(4)	2.37(5)	0.137(2)
1173	0.33(2)	0.295(8)	0.314(13)	0.016(1)
1332	0.311(15)	0.284(9)	0.276(7)	0.016(1)

Peak Energy	Peak efficiency per incident photon of detector based on:			
	$LaBr_3$	$CeBr_3$	SrI_2	CdZnTe
keV		%		
662	13.3	12.8	13.9	20.8

3.1.3 Conversion coefficients from pulse height spectra to dose for $CeBr_3$, $LaBr_3$, SrI_2 and CdZnTe based detectors.

In one study, the conversion coefficient method was used to investigate the spectro-dosemeters based on $CeBr_3$, $LaBr_3$, SrI_2 and CdZnTe [9]. Different quasi-mono-energetic sources and X-ray fields were used to cover eight different energy regions and calculate the conversion coefficients w_i. For the energy range between 148 keV and 346 keV no source was available and therefore the X-ray quality N-300 of PTB's reference fields was used. The sources and energy ranges are summarized in Table 3 and Table 4, respectively, and the results are shown in Figure 3:

Table 3 The quasi-mono-energetic sources, their γ-peaks and the corresponding energy ranges for determining the conversion coefficients w_i [9].

Source	γ-peaks used for calculation	Energy range
^{241}Am	60	40 – 80
^{109}Cd	88	80 – 96
^{57}Co	122	96 – 148
N-300	247 (mean energy)	148 – 346
^{192}Ir	468	346 – 538
^{137}Cs	662	538 – 846
^{60}Co	1173, 1333	846 – 1654
^{88}Y	1836	1654 – 2018

Table 4: The resulting conversion coefficients w_i for all detector systems (in 10^{-15} Sv, according to Eqn. 2). The uncertainties are estimated (for the expansion factor k = 1) using a Monte-Carlo method [9].

E_{mean}	w_i of detector based on			
	$CeBr_3$	$LaBr_3$	SrI_2	CdZnTe
	Sv / keV	Sv / keV	Sv / keV	Sv / keV
60	2.49(6)	1.73(4)	3.51(8)	12.0(3)
88	0.95(5)	0.86(4)	0.82(5)	5.8(4)
122	1.16(2)	1.03(2)	1.10(2)	7.2(2)
247	1.35(3)	1.36(3)	1.33(3)	10.9(3)

442	3.3(5)	3.0(5)	3.2(5)	35(5)
662	3.5(3)	3.3(3)	3.5(2)	40(5)
1250	3.45(7)	3.43(7)	3.77(7)	43(2)
1836	4.3(8)	4.0(8)	4.6(8)	53(16)

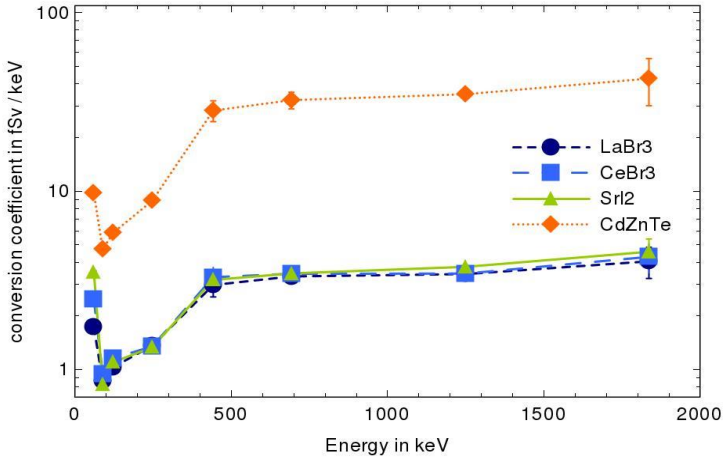

Figure 3: Conversion coefficients w_i from counts to dose. The significant higher values for the CdZnTe are due to its smaller volume of 1 cm^3 compared to 12.9 cm^3 of the scintillation detectors [22].

3.1.4 Angular dependence of the response

For the full characterization of the detector, the angular dependence of the response has to be measured [9]. This was done in PTB's reference fields in UDO II. For different energies and angles the relative response to 90° was measured, as shown in Figure 4. This has to be taken into account in case of surface contaminations of large areas, because in this case, the radiation is coming from different directions of the lower hemisphere (the hemisphere of vectors pointing from the detector towards the ground). The angles are defined so that 90° defines a plane parallel to the ground, passing through the detector, and angles larger than 90° point to the ground (i.e. to the contaminated soil).

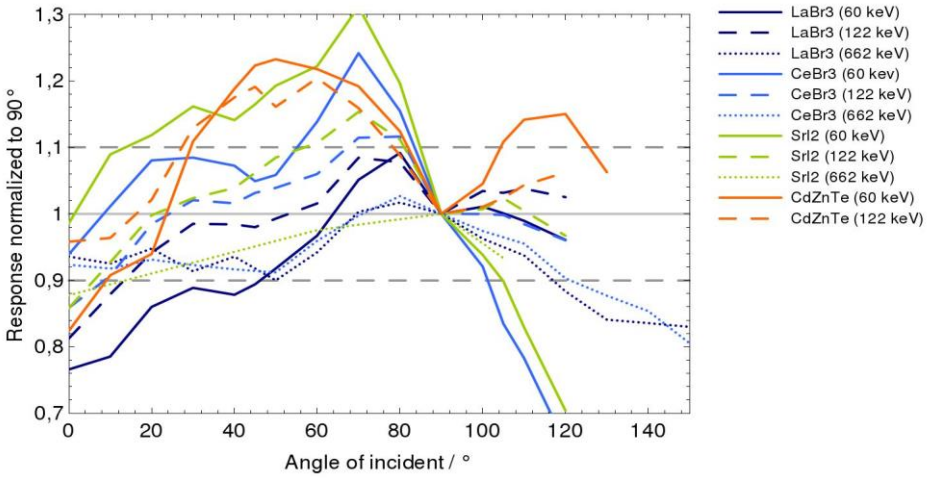

Figure 4: Relative angular response of the different detectors for 60 , 122 and 662 respectively.

The results show that for energies above 200 keV no angular dependence of the response has to be taken into account, as the deviations are less than 5%. To correct for the angular dependence at lower energies an

irradiation average angle of 105° was chosen which is the mean effective angle of radiation when having a large surface contamination in the vicinity of the detector. In a later stage of development, the housing of the detectors can be optimized. This can be demonstrated with $LaBr_3$ based detectors which is in a different housing than the other detectors and therefore shows less dependence on the photons' angle of incidence. The different shape of the CdZnTe's curve is caused by the cubic geometry in contrast to the cylindrical of the scintillation detectors.

3.1.5 Conclusion

Performance tests with the newly developed spectro-dosimetric detectors (able to measure doses and to detect which radionuclides are present) show, that these devices are suitable replacements for conventional dose rate meters, such as the Geiger Muller based dose rate meters presently used in most early warning networks. A change to these novel spectrometric detectors will bring a considerable increase in accuracy of dose rate measurements (from deviations of up to a factor of 2, improved to about 20 % or even less) of the data and, in addition, will provide nuclide specific information.

3.2 Validation of new spectrometric dose rate monitoring and contamination level estimation

3.2.1 Monte Carlo simulations

There has been considerable progress in the field of Monte Carlo simulations. Detection forcing methods have been implemented in the well-known Penelope software, and a new version of the DETEFF code, which is able to calculate the basic parameters for dose rate calculation from spectra, has been created and verified. Both computer codes were compared regarding their ability to calculate *H**(10) from spectra recorded with a LaBr3 based detector. Different methodologies were used for the calculation of H*(10). For the calculation of the detector response matrixes and the deconvolution of the measured energy loss spectra different methods were implemented and tested: a) the stripping method, b) the conversion coefficient method and c) a maximum entropy method (implemented in the code DET-H10).

The so-called 'detection forcing variance reduction' technique in open source PENELOPE software which can be combined with the other techniques already implemented, i.e., Russian roulette, splitting and interaction forcing, has been implemented to be used in environmental radioactive scenarios. The code has been tested by comparing simulations with the reciprocal variance reduction method using simple geometries.

Furthermore, MC simulations have been carried out for both detectors and different sources at UPC's secondary standard calibration laboratory using the upgraded PENELOPE/penEasy code. Spectra calculated with simulations and the experimental irradiations have been compared and analysed. There was a good agreement in the photo peak regions for the three sources, i.e., ^{137}Cs, ^{60}Co and ^{241}Am (see Figure 5). For energies higher than the photo peak energy an underestimation in the simulation can be observed. This fact is because simulation results cannot account for pile up. The sum peak of two completely absorbed photons from decay can be clearly seen for the ^{137}Cs and ^{241}Am sources. In the energy range lower than the photo peak, it can also be observed an underestimation of the simulations. In this case, the reason has a completely different explanation. The photonic irradiator consists on a roulette of sources. Therefore, it is not possible to avoid the gamma scattering as a consequence of neighbouring sources.

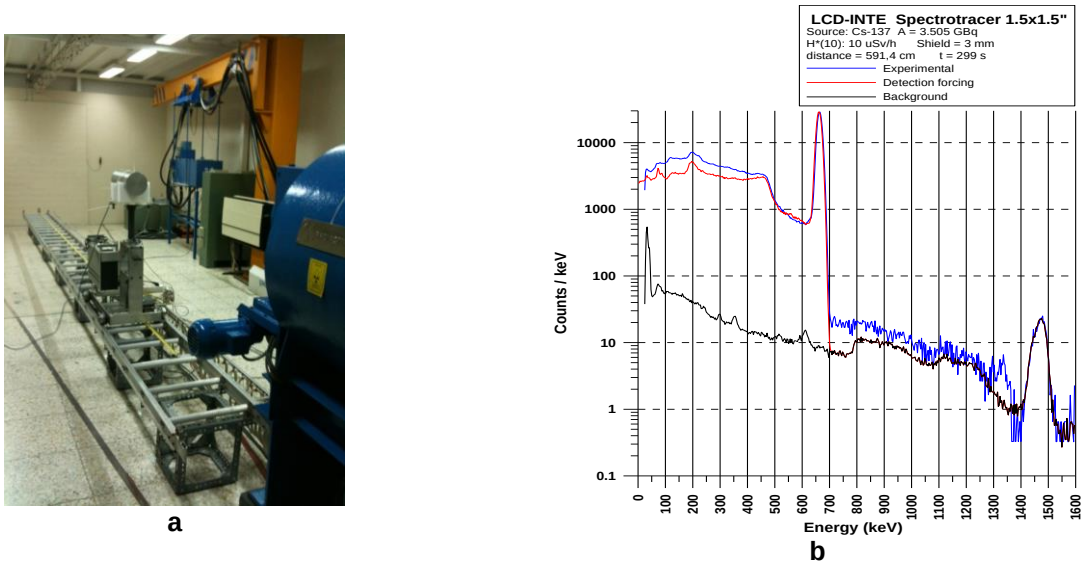

Figure 5: Picture of a) the UPC secondary standard calibration laboratory and b) Simulated and experimental spectra measured with SpectroTRACER monitor for ^{137}Cs source.

The full energy peak efficiencies for both monitors have been calculated using the MC codes developed by UPC and CIEMAT. The full energy peak efficiencies for photons of 100, 364, 1460 and 2500 have been calculated for flat surface deposition using circular geometries with radius of 5 m, 20 m, 50 m, 200 m and 1000 m. Efficiencies for semi- spherical volume clouds have been also calculated for radius of 20 m, 50 m, 200 m and 500 m. In general, the comparisons of the methodologies are in good agreement for both surface source and volume source**.**

3.2.2 Dosimetric data from spectro-dosemeters

There are several methods to calculate dose rate values from pulse height spectra. In most commercial systems, the conversion coefficient method is used because it allows the quick calculation of dose rates from measured spectra if some requirements are fulfilled. The conversion from the actual response of an instrument to a dosimetric quantity, e.g. $H^*(10)$, is made by applying the energy dependent conversion coefficients w_i to the counts of each energy bin n_i of a spectrum according to:

$$H^*(10) = w_1 \cdot E_1 \cdot n_1 + w_2 \cdot E_2 \cdot n_2 + \ldots + w_Z \cdot E_Z \cdot n_Z,$$

where E_i is the mean energy of energy region *i*. For each detector system, a full set of w_i values is derived using spectra from quasi mono-energetic sources or synthetic spectra from Monte Carlo (MC) simulations.

UPC and CIEMAT have applied stripping, conversion coefficients and Maximum Likelihood Estimation using Expectation Maximization (ML-EM) methods in order to calculate $H^*(10)$ from photon pulse-height spectra using own developed codes 'spc2dose' and 'DET-H10R'. Note: H*(10) represents the ambient dose equivalent, which is the absorbed dose equivalent (in Sievert) at a depth of 10 mm, in a virtual phantom (30 cm in diameter) of tissue equivalent material for a corresponding extended and aligned radiation field. For the purpose of validation, the methods were compared with controlled irradiations at the UPC secondary standard calibration laboratory in Barcelona, and a real scenario at the ESMERALDA station located at the CIEMAT premises in Madrid (Figure 6). In general, calculated $H^*(10)$ rates are in good agreement for the irradiations in the laboratory with relatively high dose rates compared to natural background levels. Figure 6 demonstrates that the pattern of the different methods for $H^*(10)$ rate calculations are similar and can detect both rainy periods and increased radon concentrations in air. From the comparison analysis it can be concluded that the most convenient methods are conversion coefficients and ML-EM using recalculated coefficients of the second-order polynomial energy calibration equation for each spectrum. It is advisable for the manufacturers of spectrometric monitors to improve the calibration corrections required to compensate the energy shifts due to gain changes associated with temperature variations.

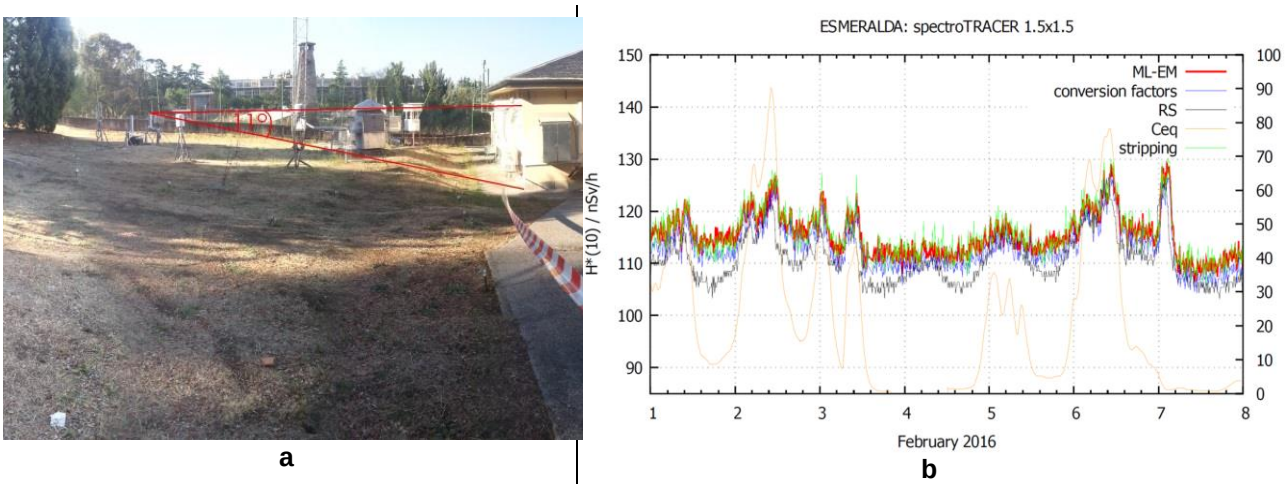

Figure 6: a) Picture of ESMERALDA reference station with the monitors installed b) *H**(10) comparison in ESMERALDA and radon progeny concentrations.

Using the developed MC variance reduction techniques, realistic spectra for fresh deposited activity of 1 kBq/m^2 have been generated for the SpectroTRACER monitor at ESMERALDA (CIEMAT) and INTERCAL (BfS) using low and high background levels, i.e, dry and rainy conditions respectively. For rainy periods, the enhanced natural peaks of ^{214}Bi and ^{214}Pb led that it would be very complicated to identify the artificial peaks, especially for ^{131}I for such deposited activities. In ESMERALDA station, due to the complexity of the geometry, which includes trees close to the monitor, depending on the deposition process on the trees the response of the monitor could be significantly different for the same grassland contamination.

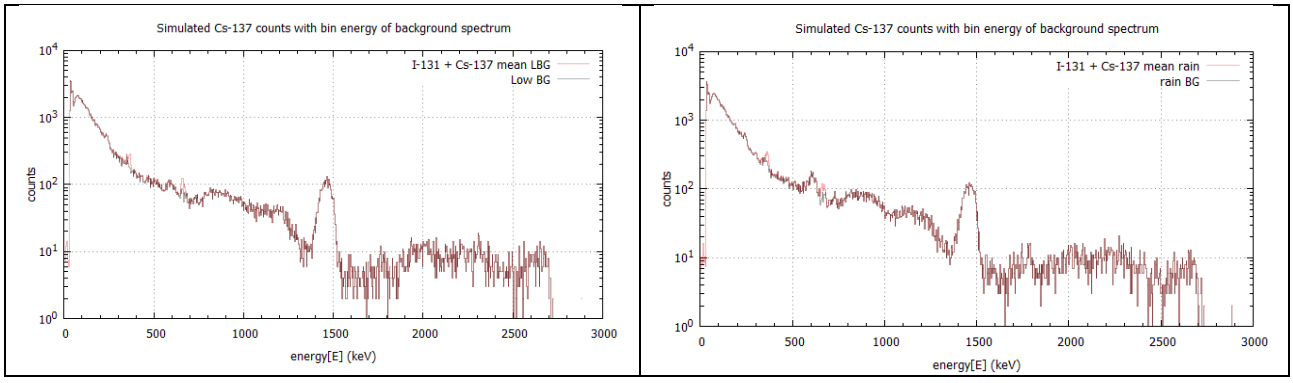

Figure 7: Detector response for fresh deposited ^{137}Cs and ^{131}I of 1 kBq/m^2 in ESMERALDA for dry (left side) and rainy (right side) scenario.

3.2.3 Results of the intercomparison of spectrometers used for dose rate monitoring

PTB performed an intercomparison of spectrometers (LaBr3, CdZnTe, etc.) to be used as dose rate monitoring instruments in early warning network systems. The important dosimetric characteristics of the spectrometers as the energy response, the linearity concerning the dose rate, the response to cosmic and terrestrial radiation, the sensitivity to detect the radiation of a radioactive plume and the temperature dependence of the indicated values were investigated. The results demonstrate that spectrometers are capable to be used as instruments for dose rate measurements, as their properties are equal or superior to conventional dosemeters.

This project investigated, as whether a new generation of spectro-dosemeters can replace conventional dosemeters, which are mainly based on Geiger-Müller tubes. Within this intercomparison, representative spectrometry systems were investigated concerning their ability to measure correct ambient dose equivalent rates. Furthermore, the data of the reference instrument, a high pressure ionization chamber, and a Geiger-Müller tube were recorded.

The measurements took place at PTB's reference measuring sites for low dose rate. The dosemeters were irradiated under reference conditions at the low-dose irradiation facility in the underground laboratory UDO II, at a floating platform on a lake (measuring site for cosmic radiation) and at a free-field irradiation facility (plume simulation machine). The resulting data contain information about the important dosimetric characteristics of the spectrometers as the energy response, the linearity concerning the dose rate, the response to cosmic and terrestrial radiation, the sensitivity to detect the radiation of a radioactive plume and the temperature dependence of the indicated values.

In this intercomparison 14 spectrometry systems were investigated. In the following, some representative results of a few of these systems are shown.

Figure 8 displays energy response curves of spectrometers developed within this project ("scientific spectrometers"), which can be compared with that of commercially available spectrometers (Figure 9, left) and conventional GM dosemeters (Figure 9, right). While the energy dependence of the response for the latter vary by a factor of up to 2 (i.e. 200%), the scientific (well characterized) spectrometers show deviations of less than 20%. Some commercially available spectrometers also show an unsatisfactory energy response, as show in Figure 9 (right).

Figure 10 depicts measured plume profiles in comparison with a reference curve. The prevailing dose rate was altered in steps by using the PTB plume simulation irratiator.

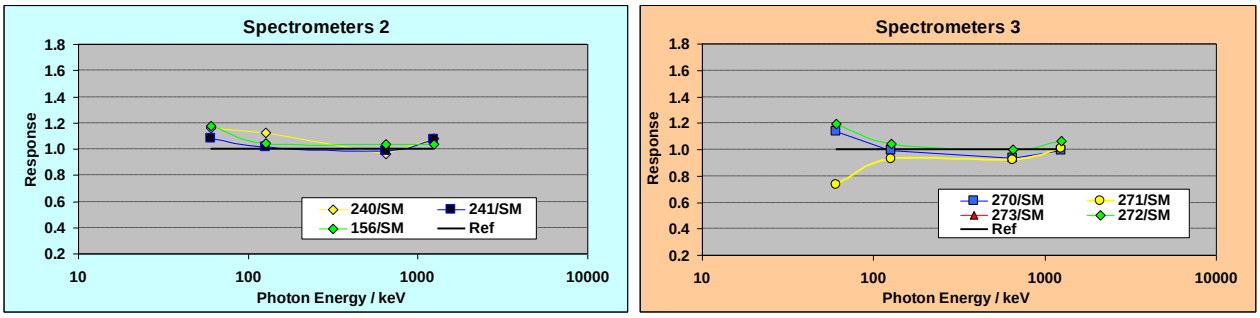

Figure 8: Energy response of eight scientific spectrometers (for comparison with Figure 9).

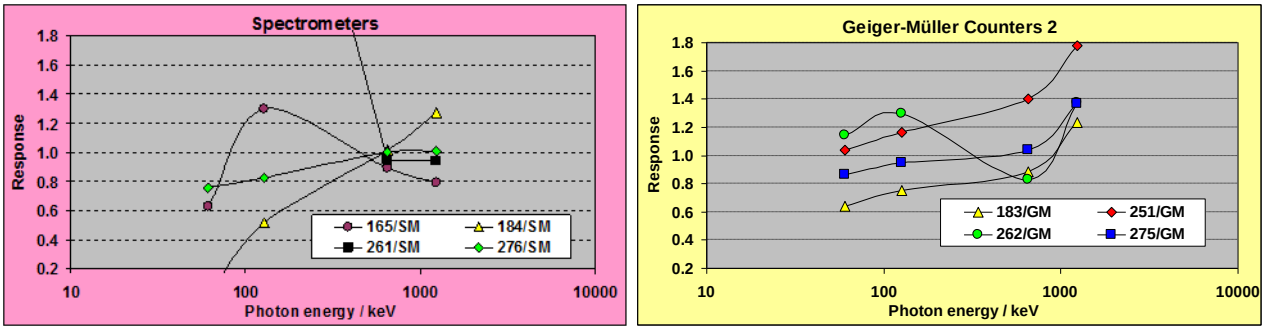

Figure 9: Energy response of commercially available spectrometers (left) and conventional GM dosemeters (right).

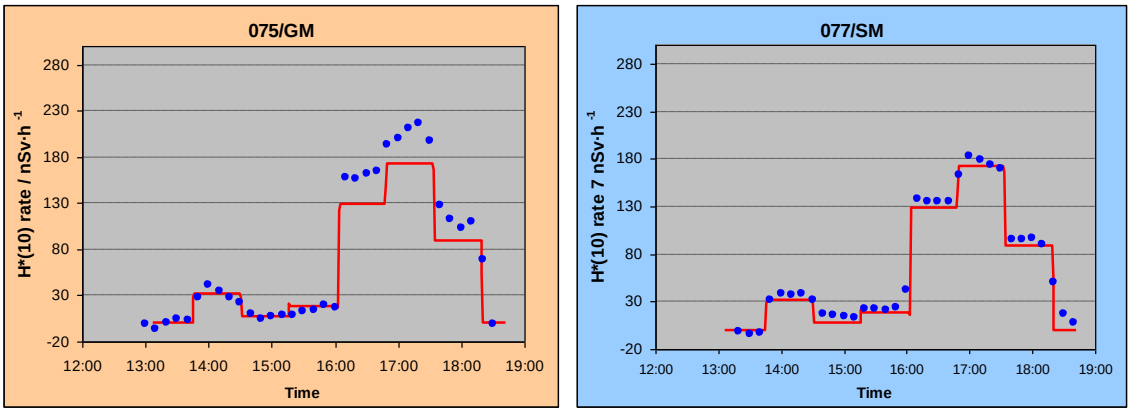

Figure 10: Measured plume profiles of a GM counter (left) and a $LaBr_3$ based spectrometer (right) after subtracting the natural background in comparison with a reference curve (red line). The prevailing dose rate was altered in steps by using the PTB plume simulation irratiator.

The results of this intercomparison show that novel spectrometers are superior to conventional dosemeters, especially to Geiger Müller counters. In addition to providing nuclide information from their spectra, their ability to measure dose rates correctly and with a high sensitivity is hardly reached by any conventional dosemeter. The energy response can be tuned to an ideal curve, just by changing parameters in the evaluation software, when the response of the detector is almost isotropic (which is fulfilled for all investigated systems). Scientific dose rate calculation software, developed within MetroERM, shows much better results than commercial software, which is not mature, yet for these measurements.

3.2.4 Conclusion

Intercomparison exercises with 15 spectrometers of 5 project partners (PTB, UPC, BfS, EHU, CIEMAT) were performed to validate the newly developed equipment and procedures for dose rate monitoring and contamination level estimation. Computer codes were compared to assess their ability to calculate H*(10) from spectra recorded with scintillation detectors. Different methodologies were used for the calculation of H*(10). For the calculation of the detector response matrixes and the deconvolution of the measured energy loss spectra; a) the stripping method, b) the conversion factor method and c) a maximum entropy method were tested. The different methods have respective advantages and disadvantages but, in general, all showed to work very well.

3.3 On-site evaluation of conditions and techniques used at dosimetric monitoring stations

It was an important aim of this project to develop scientific concepts to account for the influence of the various site-characteristics on dose rate and nuclide specific data. Methods for an appropriate correction of such data were developed and tested at some representative locations of monitoring stations. Recommendations for the selection of appropriate sites were published. Improved site characteristics will reduce uncertainties of radiological data and hence contribute to the harmonisation of early warning networks. The contributions of the different background sources to measured dose rate values of spectrometric detectors have been determined and background correction methods were developed. In addition, the results achieved have been used to provide guidance for future developments and benchmark experiments.

As a new approach in dose rate data analysis, signal processing methods were used to perform the time series analysis of long-term measurements. The results showed the typical fluctuations and their frequency, which will enter the calculation of typical uncertainties that occur during long-term dosimetric monitoring.

3.3.1 Influence of soil radon and radon progeny concentrations in air on dose rate detectors

Radon concentration as function of the soil depth was measured during the years 2011 (previous to this project) to 2015, in a location of the Aristotle University campus [8]. Radium distribution in soil was found constant. On the contrary, as expected, radon concentration increases with soil depth. The experimental distribution was reproduced by solving the general transport equation (diffusion and advection). From the general radon migration (diffusion and advection) equation, the radon exhalation rate from the soil (26.7 ± 4.5 Bq m^{-2} h^{-1}) was

indirectly deduced, from the measured radon profile in the soil. At the same location, 113 direct radon exhalation measurements were performed in the period from 2010 to 2015. From these, an average value for the six years of (21.1 ± 3) Bq m^{-2} h^{-1*} can be derived. The comparison between the radon exhalation rates deduced by the indirect and direct method indicates the validity of the diffusion advection model predictions concerning the radon exhalation rate from soil. The relation between radon migration in soil and terrestrial gamma radiation was studied. In particular, the correlation between gamma radiation one meter above soil and radon exhalation in 6 locations of the Greek early warning system network was investigated. A positive correlation between gamma dose rate in air (caused by radon progeny) and radon exhalation rate from soil was found. The rain effect on the dose rates measured by the high pressure ionization chambers (HPIC) and HPGe detectors was studied. Two remarks can be mentioned: The first is the increase of the dose rate due to air radon progeny deposition (^{214}Bi and ^{214}Pb) on the soil surface by the rain. After the air radon progeny deposition on the soil surface, the gamma dose rate decreases due to natural decay of the gamma emitters ^{214}Bi and ^{214}Pb. The second remark is that after the rain and particularly after some half-lives of ^{214}Bi and ^{214}Pb the dose rate is smaller than before the rain starts. The humidity of the soil becomes the attenuator of the gamma radiation emitted by the soil.

3.3.2 Response of early warning systems to terrestrial and cosmic radiation.

The Telemetric Early Warning System Network of the Greek Atomic Energy Commission consists mainly of a network of 24 Reuter-Stokes HPIC for gamma dose rate measurements and covers all Greece. In the present work, the response of the Reuter-Stokes HPIC to terrestrial and cosmic radiation was evaluated in comparison with spectroscopic data. The spectroscopic data were obtained by in situ gamma spectrometry measurements with portable Germanium detector (HPGe) right next to the Reuter-Stokes detectors, at six of the 24 locations of the Greek early warning system network during the period from 2014 to 2016 and at ten locations in the year 2001. The HPGe detectors are sensitive only to terrestrial gamma radiation. On the contrary Reuter-Stokes detectors are sensitive also to cosmic radiation. Therefore, the comparison between the absorbed dose rates in air measured by the two instruments can reveal information concerning the response of the Reuter-Stokes detectors to terrestrial and cosmic radiation. For the HPIC detectors a conversion factor for the measured absorbed dose rate in air to the total ambient dose equivalent rate $\dot{H}^*(10)$, due to terrestrial and cosmic component, was deduced by the field measurements.

3.3.3 Detection of rain events in radiological early warning networks with spectro-dosemeters

Short-term pronounced increases of the ambient dose equivalent rate due to rainfall are a well-known phenomenon. Increases in the same order of magnitude or even below may also be caused by a nuclear or radiological event, i.e. by artificial radiation [21]. Hence, it is important to be able to identify natural rain events in dosimetric early warning networks and to distinguish them from radiological events. Novel spectrometric systems based on scintillators may be used to differentiate between the two scenarios, because the measured gamma spectra provide significant nuclide-specific information. One study describes three simple, automatic methods to check whether an $\dot{H}^*(10)$ increase is caused by a rain event or by artificial radiation the Man Made Gross Count (MMGC) method, the peak based method and the χ2 method. These methods were applied to measurements of three spectrometric systems based on $CeBr_3$, $LaBr_3$ and SrI_2 scintillation crystals, investigated and tested for their practicability at a free-field reference site of PTB.

To test whether the three methods can distinguish between artificial and natural increases of $\dot{H}^*(10)$, simulations assuming a radioactive fallout were done. Spectra produced by the presence of the radionuclides ^{137}Cs and ^{60}Co were collected by installing weak sources of these isotopes close to the detectors at the reference site. The dose rate contributions of the sources correspond to an activity concentration of 13 kBq/m2 (^{137}Cs) and 3.2 Bq/m2 (^{60}Co), respectively. The radiation of surface activity concentrations was approximated by using point sources, which produces the same dose rate, because the detectors have a widely isotropic response (especially above some hundred), so that the origin of a photo peak in the spectrum is not of importance. Dose rate to surface contamination coefficients were applied to calculate the virtual surface contamination. As an example, the results obtained from measurements with a $CeBr_3$ spectrometer are shown in Figure 11. All results obtained by this type of investigation are listed in Table 5.

The MMGC method is surely the easiest method to be implemented. It does not require any complicated preparation, only the spectrometer's energy calibration must be known. In the selected scenarios, the presence of artificial radionuclides is detected, independently from the detector used. If this method was used to interpret gross data of a $LaBr_3$ detector, it produced a significant number of false alarms during heavy rain events,

because of artificial radiation below 1400 and the radiation of radon progeny cannot be distinguished. This problem is not observed if background corrected data are analysed instead of gross data. However, the sensitivity to detect radiological events will be reduced drastically after background subtraction because of the considerably higher variance of the data, so that this approach is not favourable.

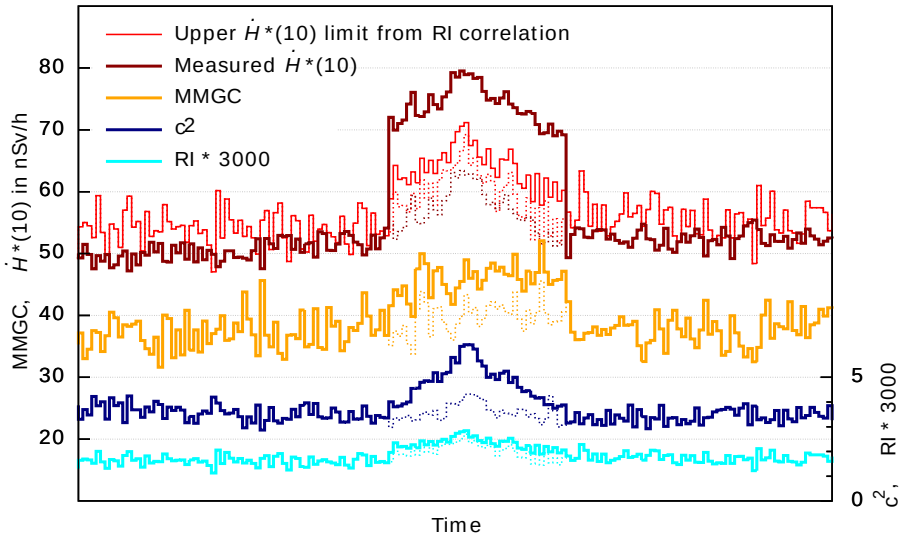

Figure 11. Results after applying the three methods for rain detection to data of a $CeBr_3$ spectrometer, by simulating a radioactive fallout by using ^{137}Cs and ^{60}Co sources during a rain event. The dotted lines show dose rate values and results of the methods without the addition of the artificial spectra [21].

Table 5. Detection of an artificial increase of the dose rate, which was ca. 20 nSv/h [21], by the different investigated methods.

Test case	Method	$LaBr_3$	$CeBr_3$	SrI_2
$^{137}Cs+^{60}Co$ during rain event detected?	peak based	yes	yes	yes
	MMGC	yes	yes	yes
	χ^2	yes	yes	no
$^{137}Cs+^{60}Co$ during dry period detected?	peak based	yes	yes	yes
	MMGC	yes	yes	yes
	χ^2	no	no	no
^{131}I during rain event detected?	peak based	-	no	-
	MMGC	-	yes	-
	χ^2	-	yes	-
^{131}I during dry period detected?	peak based	-	no	-
	MMGC	-	yes	-
	χ^2	-	yes	-

The peak based method is more difficult to implement because it requires a good knowledge of the detector's efficiency and energy resolution as the function of energy. The results concerning the selected scenarios are good as well, except for the test cases involving the ^{131}I source, which has peaks of energies similar to that of radon progeny. These two methods can also be combined to obtain results that are more reliable.

The χ^2 method turned out to be the most problematic. Under the assumptions made in this work [21], this method could correctly detect most of the rain events only if data of SrI_2 detector were evaluated, but could not distinguish artificially induced $\dot{H}^*(10)$ increases from natural rain events. However, selection of different parameters may improve the performance of this method. This has to be investigated in further studies.

3.3.4 Conclusion

Corrections for the influence of on-site characteristics on dose rate and nuclide specific data were developed and tested at monitoring stations. Recommendations for the selection of appropriate sites have been submitted for publication, including improved site characteristics that will help to reduce the uncertainty of radiological data and hence contribute to the harmonisation of early warning networks. In addition, background correction methods were developed for dose rate measurements using spectrometric detectors.

3.4 Improved detection methods and data analyses techniques for radon at low concentrations

Measurements with dose rate meters and spectrometers in known reference atmospheres of radon and its progeny in the radon-chamber of PTB revealed that low concentrations of radon in air may influence dose rate measurements only slightly. Nevertheless, it was found that a considerable part of the radon progeny may be attached to the surface of detectors. Due to the close geometry, these attached progeny have an influence on the measured results. The potential influence of radon progeny on outdoor dose rate measurements was estimated and published. The influence of radon progeny on activity concentrations measured by air-samplers was also investigated.

The time dependency of soil radon concentrations, radon progeny concentration in air, gamma dose rates and radon exhalation rates, were measured at a dedicated installation. Highest correlations of radon concentrations and dose rates have been found when the atmospheric parameters vary smoothly due to the atmospheric stability. In addition to these measurements, MC simulations have been performed and the results compared with the experimental results.

3.4.1 Influence of radon on dose rate measurements, studied in PTB's Radon chamber

If radon is accumulated in the natural environment at a dose rate measuring station, the air volume in which a higher radon concentration is found is limited, e.g. because of the topology or by inversion layers. If a volume of about 3000 m^3 is assumed (very high radon concentrations will likely be observed in the environment if the soil exhalates a high radon flux, an inversion layer exists and if the air exchange around the dose rate instrument is somehow prevented by calm air between buildings, dense vegetation, etc.), the conversion coefficient of 0.3 (nSv/h)//(Bq/m^3) would be valid according to the published results of a study [24]. As a consequence, a rise of the indicated value of the dose rate instrument of some nSv/h is possible, when very high activity concentrations of some 10 Bq/m^3 are assumed (which rarely has been observed in nature).

The radon and hence the radon progeny (i.e. the decay products of radon) activity concentrations in soil and in air depend on various geological as well as meteorological conditions and cause significant fluctuations of the normal background radiation level; i.e. the ambient dose equivalent rate in the normal environment. The influence of radon progeny on dose rate instruments was studied. Benchmark experiments were performed to compare MC simulations with measurements from a well-defined radiation field produced by Rn progeny in PTB's radon reference chamber [24]. The measurements show that the $H^*(10)$ rates measured by different monitors have a significant contribution due to the deposition of radionuclides on the surface of the monitor (plate-out).

From the measurements and MC simulation, a conclusion can be drawn that under extreme conditions, radon in air can have an effect on indicated dose rates of area dosemeters operated in the natural environment of some nSv/h or even more then 10 nSv/h under extreme conditions. However, as a prerequisite for the scenario, a number of favourable conditions have to be assumed which lead to very high and unusual radon activity concentrations, so that such conditions will rarely occur.

The plate-out contribution is different for each monitor due to different housing geometries, materials and surface texture and flaws the direct measurement of dose rates considerably. This effect has to be taken into account whenever indoor measurements of dose rates are performed in a radon atmosphere. Campaigns of indoor ambient dose rate measurements are often carried out in areas with high Rn activity concentrations such as mines, other underground working places, spas, etc. In these locations, it is usual that high Rn-progeny concentrations are present in the air, so that conditions comparable to those in the PTB radon chamber prevail. The measurements show that the direct measurement of dose rates by exposing a dosemeter are not possible, because the indicated values could be wrong by an order of magnitude, depending on the conditions. In an

outdoor situation, however, the contribution to *H**(10) of deposited radon progeny activities on the detector surface is small compared to the contribution from the gamma radiation emitted from Rn progeny in a huge air volume.

3.4.2 Blank indication of active radon monitors

Accurate determination of blank indication of radon measurement instruments (indication corresponding to a null radon concentration, sometimes referred to as intrinsic instrument background), may have similar weight as determination of the instrument sensitivity. As an example, for a radon monitor with blank of 30 Bq/m3, used to measure a typical radon concentration of 600 Bq/m3, the blank correction amount to 5 %, that is well beyond the typical sensitivity uncertainty. Active monitors can be contaminated by 210Pb and may give blank indications of the order of tens Bq/m3, if the instruments have been exposed to very high radon concentrations for long times. Additional sources of non-zero blank can be related to electronic noise and electronic setting.

In order to investigate the equivalence of their capability to determine the blank indication of radon monitors, BfS and ENEA measured independently the blank indication of an “AlphaGuard” radon monitors in their respective radon chambers. The results achieved by the two participant laboratories where submitted to an independent referee before disclosure of the individual results. The results of the two blank indication determinations were: (2.5 ± 0.5) Bq/m3 by ENEA and (2.9 ± 0.6) Bq/m3 by BfS, respectively.

The two results well agree within the reported uncertainties proving that procedures applied in both laboratories are reliable and provide the required uncertainties. It should be noted that the good agreement obtained is even more indicative considering the difficulty of this measurement, the low value of the blank indication and the special consideration that this is, to our knowledge, the first comparison of this type organized at the international level.

3.4.3 Conclusion

Dose rate and airborne radioactivity monitoring:

Measurements with dose rate meters and spectrometers in known reference atmospheres of radon and its progeny in the radon-chamber of PTB revealed that low concentrations of radon in air (in the range from 300 Bq m^{-3} and below) may influence dose rate measurements only slightly. However, it was found that a part of the radon progeny may attach to the surface of detectors and, because of the close geometry, therefore influence measurement results. The potential influence of radon progeny on outdoor dose rate measurements was estimated and found to be small (in maximum a few nSv/h) for typical outdoor radon concentrations. The influence of radon progeny on activity concentrations measured by air-samplers was also investigated and found to reduce the detection limits of some relevant radionuclides.

Radon monitoring:

There is a memory effect of radon monitors which have been exposed to high levels of radon activity concentrations, called “blank indication”; i.e. the reading of the instrument, remaining even in the later absence of any radon. This inherent background of the instrument, which has to be corrected for, especially when low radon concentrations have to be measured, is caused by internal contaminations of the instrument with long-lived radon progeny from former exposures to radon atmospheres. The blank indication of a radon monitor was investigated at radon chambers of ENEA and BfS. The results achieved perfectly agree within the reported measurement uncertainties.

3.5 Novel and improved instrumentation for airborne radioactivity monitoring

Three on-line instruments for radioactive-aerosol monitoring have been developed within MetroERM. All three instruments provide high-resolution gamma spectra for sensitive determination of nuclide-specific activity concentrations levels-in-air, in near real time. The instruments are suitable for permanent installation at a monitoring station and provide significant improvements over current off-line aerosol monitoring. A review of key results relating to these instruments is given below.

CIEMAT have developed a continuous on-line air sampler, based on a continuous glass fibre filter and an electro-mechanically cooled HPGe detector. The system has been running for six months at the CIEMAT field

site, with periods of unattended operation. This extended dataset proved that the system was capable of reliably measuring key radionuclides with an improved detection limit.

CMI and NUVIA have designed and built a new modular air sampling system for in-field airborne radioactivity measurements. The system consists of a fully automated sample changer for filters, and a shielded mechanically cooled HPGe detector. Special software algorithms were developed and tested for the subtraction of natural radionuclide contributions from the measured gamma-ray spectra and for the analysis of the results. The system can be equipped with a transportable shielding.

IJS have developed a compact portable on-line aerosol sampling gamma spectrometry system based on a CeBr3 detector. It is capable of providing continuous on-line low level airborne radioactive particulate monitoring for field station use via 3G network communications. The calibration of the device and performance tests (using spiked filters as well as exposure to environments with elevated natural radon level) were performed at NPL. Further tests performed in a controlled radon environment at ENEA confirmed the suitability of the device for its purpose.

3.5.1 CIEMAT airborne radioactivity monitoring system

The airborne radioactivity monitoring system installed at ESMERALDA, the CIEMAT´s reference site for radiological and weather measurements for continuous monitoring of airborne particulate radioactivity, consists of an electro-mechanically cooled High-Purity Germanium (HPGe) detector operated by a portable spectroscopy workstation (shown in Figure 12). Air is sampled at a flow rate of 25 $m^3 \cdot h^{-1}$ and particles are deposited onto a continuous glass-fibre filter directly exposed to the HPGe detector. An option exists for the installation of a second spectrometry detector (usually scintillation detectors such as NaI:Tl, $LaBr_3$:Ce or $CeBr_3$) at the top of the measurement chamber for direct and real time comparison with the HPGe detector.

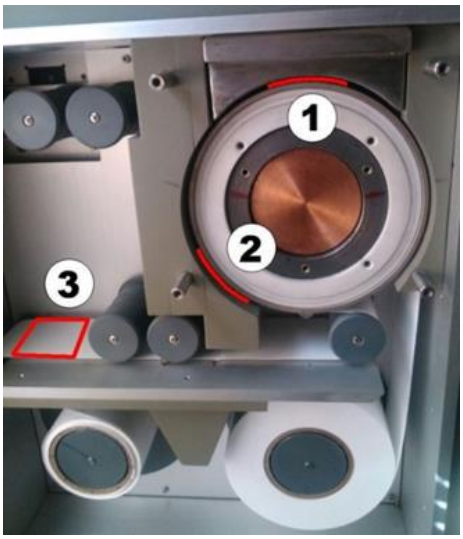

Figure 12: Automatic radioactive aerosol monitor developed by CIEMAT. Detection efficiency values have been determined for the three positions indicated in the figure to ensure the system response is understood during emergency scenarios where deposition rates may be very high.

Specific application software, programmed in Visual Basic using ActiveX controls and dynamic libraries from Genie-2K® gamma analysis software, has been developed to control the operation of the system and perform automatic spectra analysis. Algorithms for analysis have been optimized according to the results of previous studies to evaluate the influence on detector response of particle deposits located in the vicinity of the detector's field-of-view. A MySQL database has been designed for data management and storage. A screen shot of the control and analysis software is shown in Figure 13.

The system for continuous monitoring of airborne particulate radioactivity installed at CIEMAT was operated under test conditions from May to October 2016. Typically, the system worked unattended during two or three weeks in a row and the reported results were accessed on real time and remotely by the authorised users.

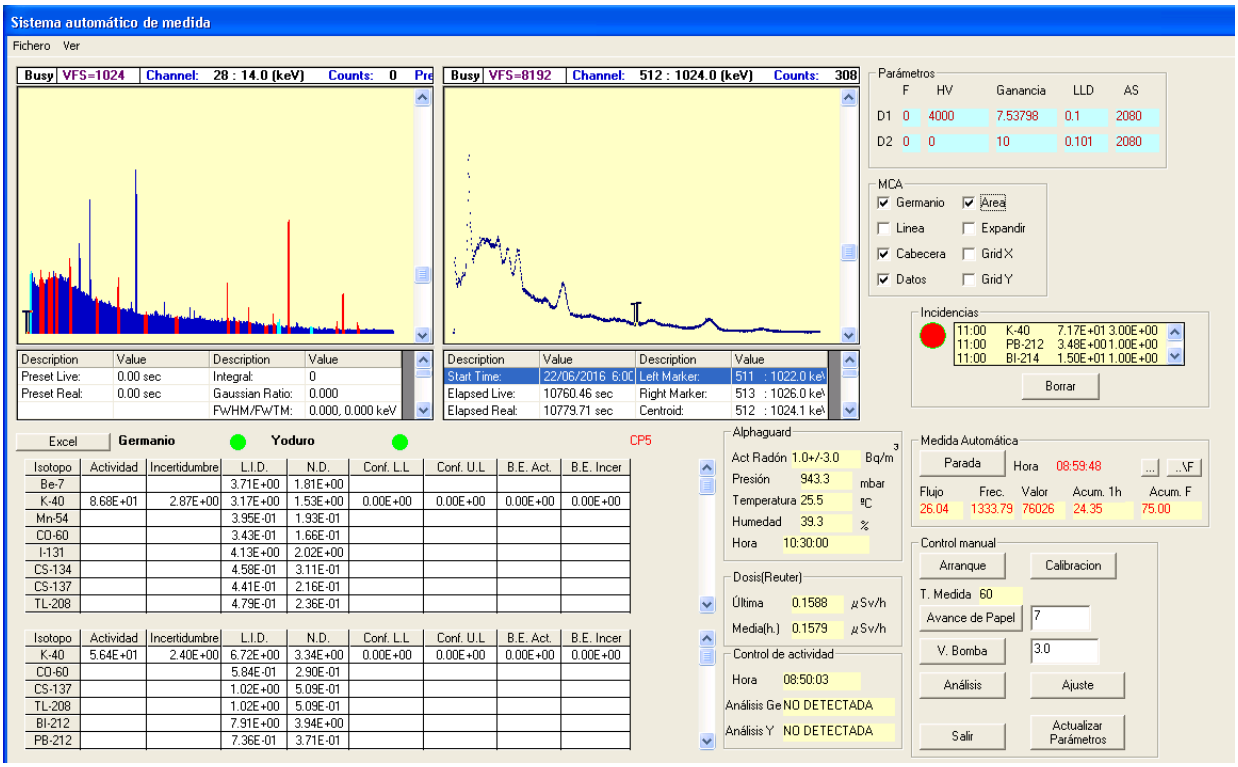

Figure 13: Graphical user interface designed for the control and analysis of results.

One of the goals for the system was achieving the detection limit values for man-made nuclides at least in the order of the required reporting levels in the radiological environmental survey programmes. For example, in Spain such levels are 0.2 Bq·m^{-3} for ^{60}Co and ^{137}Cs, and over 1 Bq·m^{-3} for short lived fission products such as ^{59}Fe or ^{65}Zn. As expected, no positive results on man-made gamma emitting nuclides were found during the 5 month period of the study. Figure 14 presents the detection limit results for ^{60}Co and ^{137}Cs during the 10 days reference period and considering four integrating periods during the routine mode: 1, 2, 6 and 24 h. Both charts show that the detection limits achieved with 1h integrating time are similar to the required reporting level of 0.2 Bq·m^{-3}. The detection limit improves with increasing integrating time and sampled air volume, achieving a detection limit in the same order of mBq·m^{-3} for the daily samples.

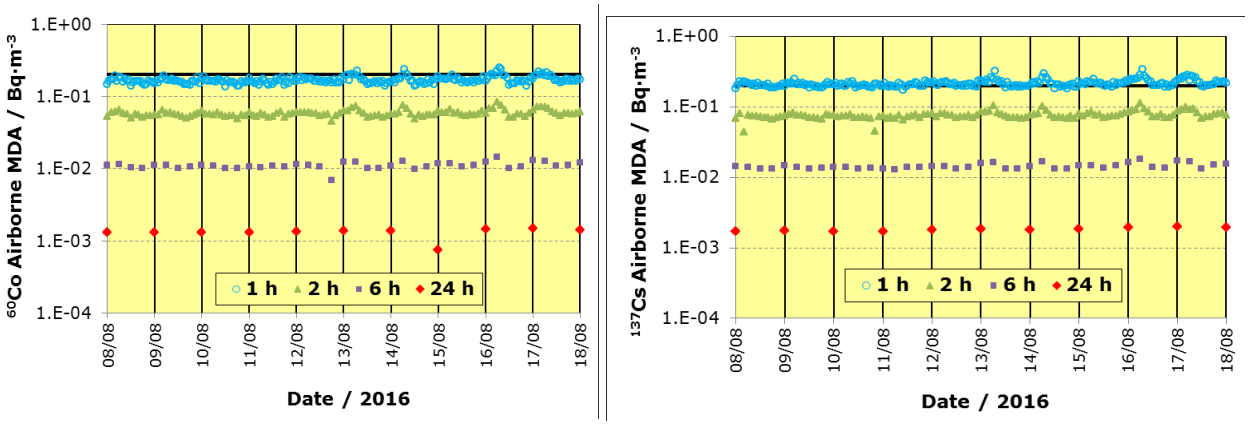

Figure 14: ^{60}Co and ^{137}Cs airborne minimum detectable activity (MDA) results obtained in the 10 day reference period for different integrating periods. The horizontal solid black line corresponds to the required reporting level of 0.2 Bq·m^{-3}.

The ESMERALDA station is CIEMAT's reference site for radiological and weather measurements. The results obtained during continued operation of 6 months demonstrate the high quality of the long-term performance of the continuous on-line airborne radioactivity measurements done.

3.5.2 CMI airborne radioactivity monitoring system

A modular, transportable system utilising an ORTEC-200V HPGe detector with a relative efficiency of 50% and a resolution of 2 keV for ^{60}Co has been developed (Figure 15). The system consists of measuring part, sampling part and a sample changer with storage for clean and used filters. The air sampling unit is based on high-power air pump capable of drawing 130 m^3 of air per hour. Air intake as well as air pipeline is heated to protect the device from condensation of water. Aerosols are filtered through filter cartridges with active diameter of 85 mm. New and used cartridges are stored in two storage tubes with capacity of 250 cartridges per tube. The advantage of this system is that filters can be analysed also after measurement in the laboratory if needed. The monitor may be remotely controlled and technical intervention is required only for replacement of the filter cartridges. This means the device may operate without service intervention for up to 250 days. The monitor is intended to be mounted within an ISO container, which can be, in addition, tiled from inside with low concrete shielding material. The system is shown in the figure below.

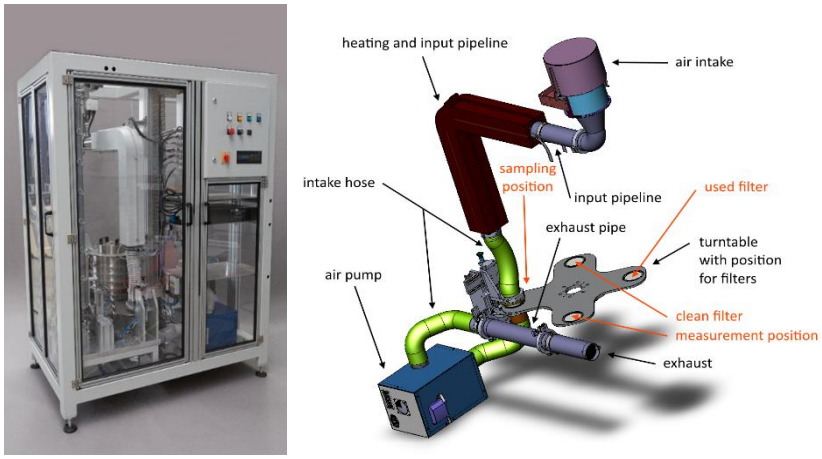

Figure 15: Modular, transportable radioactive aerosol monitor developed by CMI and NUVIA.

A Monte Carlo model of the detector assembly (Ge crystal, mount cup, and end cap) was created based on the confidential detector diagram and a detector radiogram obtained at CMI. The model was validated by comparison to experimental measurements. Full energy peak detection efficiency (FEP DE) values were measured for gamma-ray emitting point sources positioned on the detector's rotational axis at the distance of 25.1 cm from the detector end cap. Three identical detectors were tested. The sources used in the measurements were added into the MC model of the detector. The source distance from the detector end cap was set to be the same as in the experiment. The relative differences between the simulated and measured FEP DE values are plotted in the Figure 16. Excellent agreement was found between the simulated and measured data for all three detector units.

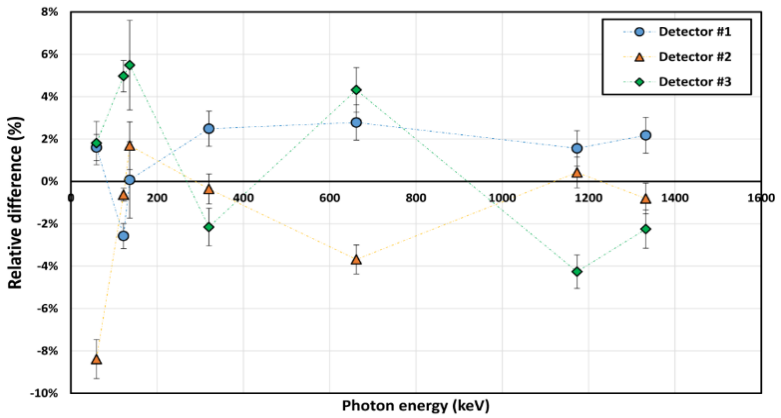

Figure 16: Comparison of simulated and measured FEP DE values for gamma-ray emitting point sources located at a distance of 25.1 cm from the detector end cap.

3.5.3 IJS aerosol sampling and measuring device A compact, portable aerosol sampling and measurement device (Monitoring Air pump for Radioactive aErosols, MARE) with a $CeBr_3$ scintillation detector positioned centrally within a concertinaed filter assembly and an improved high flow rate air pump was developed by IJS. It provides continuous on-line low-level airborne radioactive particulate monitoring for field station use via network communications.

The prototype device consists of three main units: detection, air and base unit. A $CeBr_3$ scintillation detector was selected due to its low intrinsic background radioactivity and very high light output and linearity, which results in good energy resolution. A digital signal processing device was developed in-house. The air unit consists of a concertinaed aerosol filter assembly, a flow meter and a high-performance air pump with a stable flow rate up to 200 m^3h^{-1}. The base unit consists of a microcontroller unit with a colour touch-screen 800×480 TFT display supporting a graphic user interface (GUI) for easy interaction with the system in the field, or via a GPRS/3G connection. All measured parameters and system settings are controlled by this unit. It also supports a connection to an external computer to perform heavier real time computing, to make other decisions and to communicate and send data to the user. The MARE device is presented in the figure below.

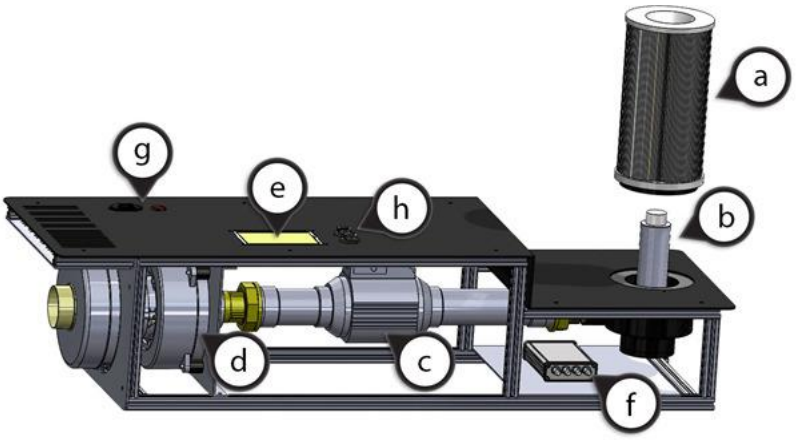

Figure 17: Novel portable Monitoring Air pump for Radioactive aErosols (MARE). The systems constist of the following components: a) concertina shaped aerosol filter, b) $CeBr_3$ scintillation detector with photo multiplier read-out, c) air-flow meter, d) air-pump turbine, e) microcontroller unit with touch screen, f) preamplifier and digital pulse processing unit, g) 230 V power socket, h) USB sockets for communication with a PC.

The validation of the compact radioactive aerosol particulate monitoring device was performed at NPL. Two filters were spiked with a certified solution containing a set of radionuclides with 14 prominent gamma-ray lines between 60 keV and 1836 keV. The activities of the radionuclides in the mixture were chosen so that the

relative intensities of the peaks in the gamma-ray spectrum were comparable (as of the reference date). To make the distribution of activity over the filter as homogenous as possible, filters were spiked by drop-deposition of the solution on the filter cylinders along 11 equidistant rings. A total of 47.5 kBq was deposited to produce a high activity filter and 153 Bq for a low activity one. The measurement with the high activity filter was performed by IJS at NPL during approximately 1h without pumping air through the filter. The gamma-ray spectrum collected from this measurement (shown in Figure 18) was used in calculation of the photo-peak efficiency calibration curve, since the exact activities of each radionuclide spiked on the filter are known a priori. The total photo-peak efficiency is slightly less than 1 %, consistent with the size of the detector crystal and the average distance from the filter paper. The efficiency curve was additionally checked with measurement in a room with elevated level of radon concentration at NPL. On the basis of results of this measurement it was clear that radon progeny affected the MDA. Under typical conditions at field station use, only at areas with high radon concentrations in air, this effect has to be considered.

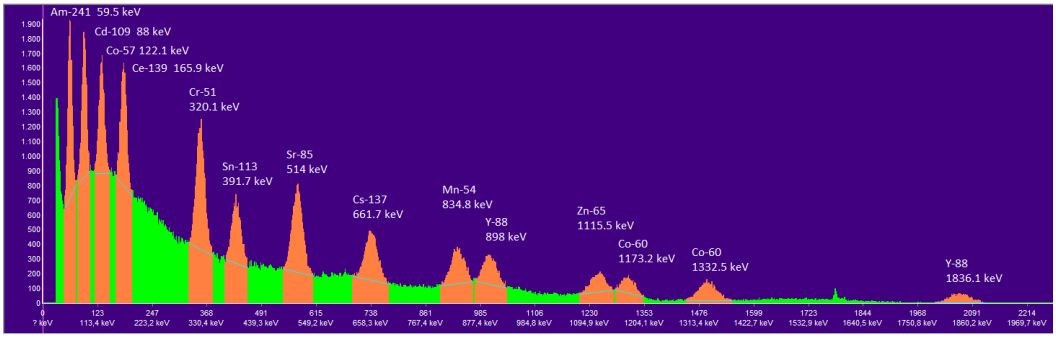

Figure 18: Energy spectrum of a high activity filter measured for 3,440 seconds.

3.5.4 Conclusion

Three novel prototypes of portable aerosol monitoring systems were set up by CIEMAT, CMI and IJS: CIEMAT developed a continuous on-line air sampler, based on a continuous glass fibre filter and an electro-mechanically cooled HPGe detector. CMI and a Researcher Excellence Grant (REG) at NUVIA designed and built a new modular air sampling system for in-field airborne radioactivity measurements. The system consists of a fully automated sample changer for filters, and a shielded mechanically cooled HPGe detector. In addition, the system can be equipped with a transportable shielding against external radiation (which would affect measurements and reduce sensitivity). IJS developed a compact portable on-line aerosol sampling gamma spectrometry system based on a CeBr3 detector.

A comprehensive comparison study of these novel spectrometers with HPGe (traditional lab based detectors), CdZnTe and CsI (a new type of scintillation detector) detectors was performed at NPL. The range of spectrometers selected represents new and emerging technologies for radionuclide detection. The results of this study and that of a laboratory comparison exercise with 66 participants, using “spiked filters”, allow national networks to decide which technology is most suitable for their individual needs, and how best to apply it. However, the results of the laboratory comparison exercise for ^{131}I-activity in air, showed unexpected issues (systematic underestimations of the 131I-activity) which requires further investigations and appropriate precautions in case of a real emergency.

3.6 Validation of new techniques in airborne radioactivity monitoring

Similar to dose rate monitoring), novel spectrometric detectors with medium energy resolution such as LaBr3, CeBr3 and SrI2 have been investigated experimentally as well as by various Monte Carlo simulations for their capability to analyse activity concentrations of radioactive particulates collected at the filters of air samplers.

A comprehensive comparison study of these novel and emerging spectrometers that have the potential to improve radioactivity-in-air measurements for national monitoring networks was completed at NPL. Five

detectors were chosen for investigation; $LaBr_3$, CeBr3, SiPM-CsI, CdZnTe and electromechanically-cooled HPGe. These detectors represent the full range of the price-performance matrix. The results achieved allow national networks to decide which technology is suitable for their individual needs and how best to apply it. Comparisons were made of energy resolution, detection efficiency and minimum detectable activity by exposing each detector to a mixed radionuclide source drop-deposited across a filter. Other factors, such as internal radioactivity, linearity, size and cost were also considered. The results have been published in a peer reviewed conference proceeding [18].

A key result from this work was the comparison of MDA values for three key radionuclides; ^{241}Am, ^{137}Cs and ^{60}Co, which are plotted in Figure 19. The HPGe and $CeBr_3$ detectors performed best, with MDA values for 24 r measurements of 0.1 and 0.2 Bq, respectively. It would take just 100 and 200 s, respectively, for these levels of contamination to be collected on a filter in an environment of 0.03 $Bq.m^{-3}$, even with a modest pump rate of 10 $m^3.hr^{-1}$ (typical of a handheld air sampler). The MDA values of CdZnTe were found to increase with energy as detection efficiency decreased. The MDA values of $LaBr_3$ were found to decrease with energy. This can be explained with reference to the internal background, which decreases from 60 to 1332 . The MDA values produced by the CsI detector were reasonable despite the poor energy resolution.

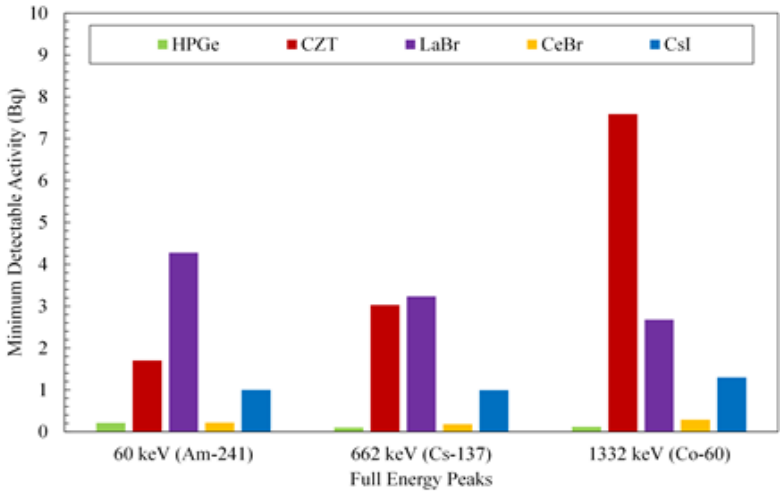

Figure 19: Comparison of MDA values for ^{241}Am, ^{137}Cs and ^{60}Co when measured in Cu/Cd lined Pb coffin over 24 h.

3.6.1 Subtraction of the natural radiation contribution to HPGe gamma-ray spectra

A method has been developed by CMI for the subtraction of Monte Carlo simulated gamma-ray spectra of natural radiation from measured spectra aiming to decrease the decision threshold for the detection of artificial radionuclides. HPGe detector spectra were simulated for selected naturally occurring radionuclides deposited onto a filter used for aerosol collection in a newly developed airborne radioactivity monitoring system. Stepwise, the simulated spectra were fitted to the actually acquired gamma-ray spectrum and the decision threshold was determined. The contribution of cosmic rays to the background was also estimated and removed.

Two examples are shown in Figure 20, where simulated spectra of several naturally occurring radionuclides have been fitted to the measured spectrum and then subtracted. A 14 hours measurement following one hour of ambient air sampling (at 60 m^3h^{-1}) is presented in the top plot. The final subtracted spectrum in this plot displays a background reduced by almost two orders. The two remaining peaks are 1) the 511 keV annihilation peak and 2) the 1460 keV ^{40}K peak. The plot on the bottom shows the combined spectrum of two measurements; firstly a 100 s measurement following one hour of air sampling (at 40 m^3h^{-1}), and secondly a 400 s measurement of a “clean” filter spiked with 10.0 Bq of ^{137}Cs. This plot is centred on the ^{137}Cs 662 peak and shows the efficient removal of the 665.6 keV interference peak originating from natural ^{214}Bi. Removal of this peak improves the ^{137}Cs decision threshold by 38%.

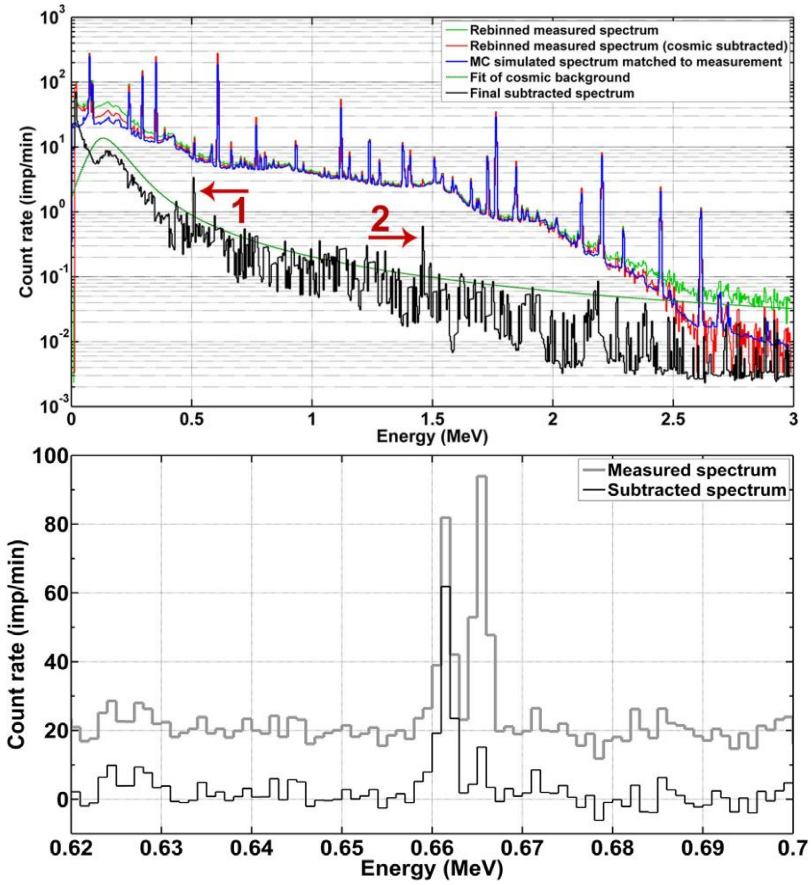

Figure 20: Top: Subtraction of natural radionuclides ^{214}Bi, ^{208}Tl, ^{214}Pb, ^{212}Pb, ^{212}Bi, and cosmic rays contribution from measured spectrum and (bottom) region of interest around 662 keV peak of ^{137}Cs. Subtraction of background removes the ^{214}Bi 665.6 keV peak near the ^{137}Cs peak and improves measurement sensitivity.

3.6.2 Measurement of natural background made by CIEMAT

The unavoidable contribution of natural gamma emitters, mostly ^{40}K (1460 main line) and ^{214}Bi (609 main line), to the collected spectra affects the performance of a system to determine airborne radioactivity, in particular to the minimum detectable activity. Their influence needs to be considered before studying the capabilities to detect man-made nuclides. The CIEMAT aerosol monitor was operated over a 6 month period and used to measure the concentration of the key natural radionuclides of ^{40}K and ^{214}Bi. The results for a 10 day period are given in Figure 21.

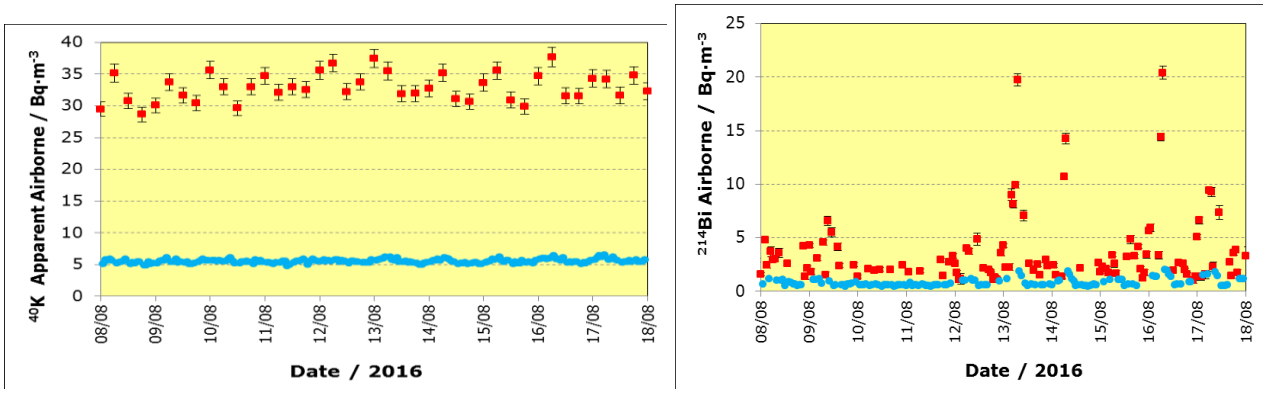

Figure 21: Hourly ^{40}K (left) and ^{214}Bi (right) airborne activity concentration results for a ten day reference period.

The red squares in the ^{40}K plot show the 'apparent' concentration values with their corresponding uncertainties (k=1) as error bars, that is, 'positive measurements', understood as clearly over the MDA. In all cases, the values correspond to the first measurement in each 6 h cycle, i.e. without any background subtraction. The values are approximately constant (within 10%). The blue circles correspond to the subsequent spectra in the 6 h cycle, which were obtained by subtracting the precedent spectra as background. In these cases, the 40K 'apparent' airborne activity concentration was below the MDA. This fact clearly indicates that the 1460 keV signal in the spectrometer was mainly due to the materials surrounding the detector, but was not coming from the sampled air particulate matter.

The red squares in the ^{214}Bi plot indicate the 'positive' measurements, with their associated uncertainties (k = 1). Where a positive result was not obtained, the MDA value is reported instead (blue circles). By contrast with the ^{40}K results, significant and consistent changes have been observed for the ^{214}Bi airborne activity concentration, being sensitive enough to detect activity concentrations of 1 $Bq \cdot m^{-3}$ with 1 h sampling time, and providing spectrometric analysis after a matter of seconds. Furthermore, the observed time pattern closely follows 'daily radon progeny cycle' due to radon exhalation from the soil, with a maximum early in the morning to a slight decrease during the day.

3.6.3 Measurements made within ENEA's radon chamber

The study of the performance and response of the IJS aerosol monitor to the natural background at controlled conditions with elevated radon concentrations was performed in a walk-in radon chamber at the National Institute of Ionizing Radiation Metrology of ENEA. In this chamber, elevated radon concentrations are controlled via a global ventilation system and controlled exchange of radon-rich air. In parallel with measurements of radon progeny with aerosol monitoring device two AlphaGUARD radon monitors and Tracerlab Radon Daughter Monitor were used. Three measurement campaigns were performed: a) at a high radon concentration of about 3000 Bq/m3 without production of artificial aerosols, b) at the same radon concentration with artificial aerosols generated by candle burning, c) at an intermediate radon concentration of about 1000 Bq/m3 without artificial aerosols.

The results of the measurement of the ^{214}Po alpha spectra measured by the Tracerlab instrument were compared to the measurements of ^{214}Bi (from the 609 keV gamma-ray emission) measured with the IJS aerosol monitoring (see Figure 22). On the basis of this comparison the filter efficiency of the IJS aerosol sampling device for the specific aerosol characteristics was estimated to be (88 ± 24) %. Radon progeny concentration in the environment results in an increase of the MDA in specific gamma-ray energy regions. To make the system more sensitive to longer-lived airborne radionuclides it is recommended to periodically turn off the pump for two to three hours to allow the total decay of radon progeny. These tests confirmed that the device, developed at IJS, is suitable for continuous on-line low level airborne radioactive particulate monitoring for field station use.

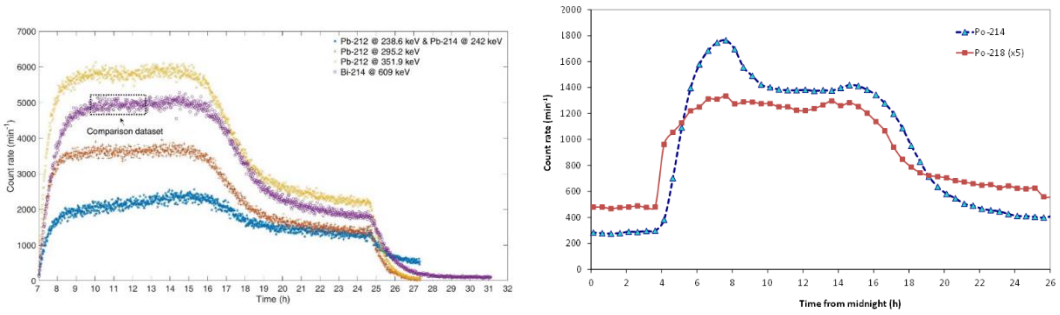

Figure 22: Comparison of radon / thoron progeny attached to artificial aerosols measured with the MARE device (left) and the Tracerlab instrument (right).

3.6.4 Fast radiochemical analysis methods

This project developed faster and improved radiochemical methods for the determination of alpha and beta particle emitting radionuclides (Sr-90 and U, Pu and Am isotopes) in aerosol samples collected using high

volume air samplers, which can only be detected after a radiochemical separation and purification procedure. Such radionuclides are regularly measured within environmental monitoring networks, but the procedures commonly used require weeks to complete the analyses, which makes them inadequate for emergency situations. In order to enable the determination of such alpha or beta particle emitting radionuclides in aerosol samples gathered in routine measurements in a faster way, less time-consuming sample treatment strategies have been tested and radiochemical separation procedures have been modified to carry out simultaneous separation of the radionuclides of interest. Two radiochemical separation schemes have been developed, compared and validated using aqueous solutions, mineral reference materials and a series of real samples.

The set of methods developed shows good performance in the separation of the radionuclides and a significant reduction of the time of analysis, which would allow reporting increased activity concentrations of alpha and beta particle emitting radionuclides during a radioactive plume deposition. PTB and TAEK have participated in these developments. The purpose of the work was to assess and compare the performance of two analytical methods (A and B) for the simultaneous radiochemical separation of americium, plutonium, uranium and strontium in air dust samples, which could be employed in emergency situations. The two methods are based on two different arrangements of extraction chromatography resins for the radiochemical separation. The methods were tested with aqueous solutions and with different types of mineral samples. The results have shown that both methods perform well and can be used to obtain valuable radiological information in a short time.

The tracer recovery values of the radiochemical separations of a soil reference material are presented in Figure 23. Recovery values over 50 % were achieved in all cases. Recovery values for uranium and strontium are similar for both methods, but in the case of americium and plutonium, recovery values of Method B are about 20 % higher, which indicates a different performance from what was observed in previous tests using aqueous solutions.

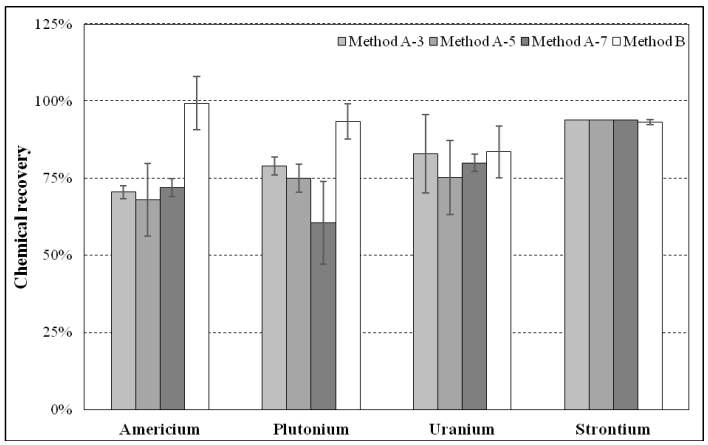

Figure 23: Mean recovery values obtained for Method A and B with a soil reference material

The results of the activity concentration determinations show good performance in the case of uranium and plutonium isotopes, which present bias values lower than 20 % in most cases. In the case of americium, Method A shows good performance while Method B presents a bias of 45 %, although recoveries for this element were high and the concentration is over the minimum detectable activity. Finally, in the case of strontium, Method B showed good performance, while bias for Method A resulted to be lower than 30 % only when 3 g of sample were taken for the analysis.

3.6.5 Conclusion

A method was developed for the subtraction of Monte Carlo simulated detector spectra of the natural background radioactivity from measured detector spectra. The aim of the method was to decrease the Minimum Detectable Activity of artificial radionuclides. After subtracting simulated spectra for all relevant naturally occurring nuclides and an estimation of the background caused by cosmic rays, the final spectrum included only the contribution of un-subtracted radionuclides resulting in significantly reduced Minimum Detectable Activity values. Newly developed equipment and procedures in the field of airborne radioactivity measurements were tested at reference installations of the project partners. All three novel portable aerosol

monitoring systems, described in section 3.5. showed their capability to detect low activity concentrations of artificial radionuclides in air.

3.7 Common metrological procedures and implementation of traceable calibrations

The metrological foundation of dosimetric measurements in the environment as well as the traceability of calibrations of activity concentration measurements in air were important aims of this project. Traceable calibrations and measurements are a prerequisite for the harmonisation of the European early warning networks. The implementation of proposed harmonised methods in this field will significantly reduce uncertainties in radiological data from typically a factor of 2 or more to a level of 30% to 40% for dosimetric data (at least under reference conditions) and to uncertainties of less than 20% to 50% for off-line air-sampling measurements of radioactivity for the most relevant radionuclides released during a nuclear power plant accident. This improvement of accuracy will also be achieved by using the novel detector systems developed within the project.

On the basis of existing relevant standards, guidelines, operational recommendations and responses to the questionnaires, common metrological procedures for the harmonization of radiological early warning networks in Europe were developed and published in scientific papers and online e-learning modules.

PTB evaluated the results of two intercomparisons of dosemeters (conventional dosemeters 3.7.1 and spectro-dosemeters 3.2.3) used within dosimetry network systems in European Member States. From these intercomparisons, the basic metrological parameters of the detectors including information on the calibration could be derived to serve as input for further harmonisation procedures. In addition, the sensitivity of the dosimetry systems was examined so that conclusions can be drawn regarding the ability of the instruments to detect small changes of the dose rate.

The long-term performance of various dose rate monitors and spectro-dosemeters were tested under real weather conditions at the reference sites of CIEMAT (“ESMERALDA”; located in Madrid; see 3.2.2) and BfS (“INTERCAL”, near Freiburg, Germany, see Figure 24).

Figure 24: BfS reference site INTERCAL at the mount Schauinsland, near Freiburg, Germany.

3.7.1 Intercomparison of dosemeters used within the dosimetry network systems

An intercomparison of conventional dosemeters used within dosimetry network systems in European Member States was performed at PTB in June 2016. From this intercomparison, the basic metrological parameters of

the dosemeters could be derived, such as the inherent background, the response to various photon fields and the response to secondary cosmic radiation. In addition, the sensitivity of the dosimetry systems was examined so that conclusions could be drawn regarding the ability of the instruments to detect small changes of the dose rate. Information on the ability of the dosemeters to be suitable to be installed in European early warning network systems can be concluded.

PTB examined 15 recent dosimetry network systems, in particular those to be installed in new EU Member States, in a dedicated intercomparison. In June 2016, the measurements took place at PTB reference measuring sites for low dose rate. The dosemeters were irradiated under reference conditions at the low-dose irradiation facility in the underground laboratory UDO II, at a floating platform on a lake (measuring site for cosmic radiation) and at a free-field irradiation facility.

Participants from seven countries, amongst them network operators from five countries, brought their dosemeters to PTB for testing. In this intercomparison, basic metrological parameters of the dosemeters were determined, such as the sensitivity, the response to various photon fields and the response to secondary cosmic radiation. The ability of the dosimetry systems to detect small changes of the dose rate, such as is produced by a contamination of a few kBq m^{-2} was examined by using a dedicated machine ('plume simulator'). The results served to characterize the performance of the dosemeters to be installed in European early warning network systems. In addition, the calibration of the network operators was traced back to PTB's primary standards. Examples of the manifold results are shown below. Response curves as plotted in Figure 25 show that for some instruments the energy dependence of the response is considerable (up to a factor of 2) and hence the dose rate reading will strongly vary, depending on the respective (but typically unknown) photon energy, leading to very high uncertainties . Figure 26 demonstrates that conventional dosemeters tend to have an over-response to secondary cosmic radiation, while spectrometers have a strong under-response to this type of natural radiation.

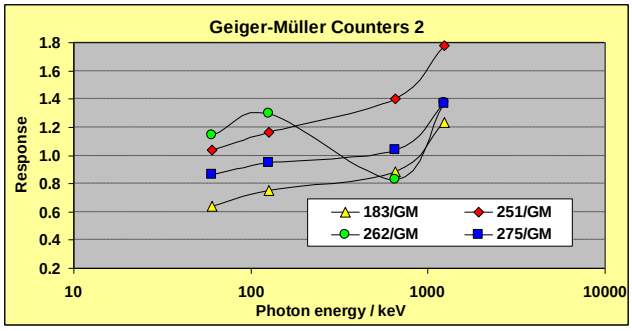

Figure 25: Example 1: Energy response of four GM counters and of four commercial spectrometers.

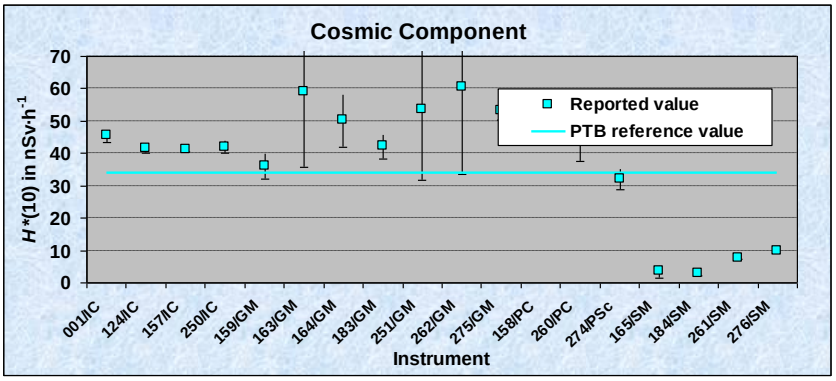

Figure 26: Example 2: Response to secondary cosmic radiation of all tested instruments.

The results of this intercomparison underlined the known disadvantages of conventional dosemeters, especially Geiger Müller (GM) counters, in comparison with more advanced dosemeters and spectrometers. Many commercial GM counters are too insensitive to be used for environmental monitoring. In contrast,

especially high pressure chambers are able to detect small changes in the environmental radiation level well. But spectrometers can be used in a way that they show a very high sensitivity combined with a very good energy response, so that they allow early warning networks precise dose rate measurements, combined with information on the nuclide vector. As observed in the spectrometer intercomparison, scientific dose rate calculation software, developed within MetroERM, shows much better results than commercial software, which is not mature or even producing misleading results, at the moment. However, the energy response of the spectrometers can be tuned to an ideal curve, just by changing parameters in the evaluation software.

3.7.2 Recommendations to harmonize European early warning dosimetry network systems

Technical requirements and methods based on realistic technical approaches and procedures tested within the MetroERM project were published to effectively harmonize area dose rate monitoring data [20]. Area dose rate monitoring has the purpose to inform about scale and intensity of any radioactive contamination in case of a nuclear emergency. In Europe about 5500 early warning stations are installed and continuously operated. Data from these stations are transmitted to the EURDEP platform, providing a close to real-time picture of the radiological situation in Europe. As a further application of early warning networks, available dose rates data from routine monitoring contain information on spatial and temporal variations of radon flux density and on soil moisture which depend in similar ways on geological and meteorological factors as the terrestrial component of the dose rate, which can be used by the scientific community for the optimization of models to determine effects of climate change.

The recommendations developed within this project addressed the following topics [20]: technical requirements for dose rate detectors installed in early warning networks, technical requirements for spectrometers, station properties of stations in networks, network strategies and topologies, data handling, station specific analysis, data transmission, central data processing, techniques and algorithms for data validation, the alert chain and alarm raising, subsequent data validation, reporting to EURDEP and data exchange, delineation on the basis of dose rate data and quality Assurance and improvement of dose rate measuring systems.

3.7.3 Traceable calibrations of early warning airborne radioactivity monitoring systems

JRC organised a interlaboratory comparison (ILC) exercise of air-sampling systems aimed at the operators of European national monitoring networks for the detection of airborne radioactive contamination both for routine and emergency response. JRC prepared spiked filter sources, containing the radioactive isotopes ^{137}Cs, ^{134}Cs and ^{131}I for this comparison and despatched them to the participants. In total 66 laboratories from 29 countries participated in this ILC. 61 laboratories are from 26 EU Member States, accompanied by 2 from Norway, 1 from Switzerland and 2 from Turkey. participated in this intercomparison in spring 2016.

All 66 participating laboratories reported valid results and in the majority of the cases these results are also reliable (i.e. the differences from the reference values were within the ±20% range). Only the evaluation of the performance of the laboratories on ^{131}I turned out to be complicated, due to two effects. First, in a number of spiked air filters a fraction of the ^{131}I activity was transferred to the protective plastic bag. Second, the integrity of the ^{131}I activity content of many spiked air filters was compromised to a variable degree. These two effects may result in a significant underestimate of the ^{131}I concentration in air. This is a very important finding, which certainly will cause further investigations beyond this project, because ^{131}I is one of the most radiologically relevant radioactive nuclides in air, released after a nuclear accident.

Reference filters for the validation of Monte Carlo detector models

Reference filters were used to validate the Monte Carlo model produced to determine the detection efficiency of the CIEMAT aerosol monitor. Three different positions of the source (8.5 × 5.5 cm^2 rectangular deposition area) were considered to evaluate the influence on detector response of particle deposits located in the vicinity of the detector's field-of-view. Figure 12(see section 3.5.1) displays these three positions. Absolute detection efficiencies for these three positions were obtained using the MCNPX V.26f code. Energy spectra were obtained for each source position and photon energy. From these spectra the full energy peak efficiencies (FEPE) were calculated as the quotient of the number of counts in the full energy peak and the number of isotropically generated photons. The simulation results were verified experimentally using a spiked glass fibre filter reproducing the rectangular deposition area. This filter was spiked with a certified solution containing a mixture of gamma emitters in the energy range from 88 keV to 1332 keV. Full energy peak areas were determined from

the spectra obtained in two source positions. Good agreement was found between the simulated and experimental efficiency values; see Table 6.

Table 6. Comparison of simulation and experimental results

Energy	Simulated values $FEPE_2/FEPE_1$	Experimental values $Counts_2/Counts_1$	Deviation %
661.37	1.23E-01	1.27E-01	-3.21
834.53	1.97E-01	1.91E-01	2.77
1172.99	3.05E-01	3.23E-01	-5.40
1332.38	3.40E-01	3.56E-01	-4.45

To validate the experimental and simulated results produced by CMI during the calibration of the modular aerosol monitor, NPL provided filters spiked with standardised solutions of gamma-emitting radionuclides (see Figure 27). Measurements of the filters were made during a collaborative test campaign at NUVIA, Trebic, attended by CMI and NPL. Good agreement was found between the three datasets, with % differences to the NPL values below ±10% across the full energy range (Figure 27). There appeared to be a systematic discrepancy between the experimental NPL values and the CMI experimental values, with an average difference of +5.5%. A small difference of just 0.5 mm in the detector-source distance (measured to be 18.075 mm) would account for this discrepancy, assuming a $1/r^2$ relationship. Such a small difference may feasibly occur if the filter is not flat in its mounting, and develops a concave or convex surface.

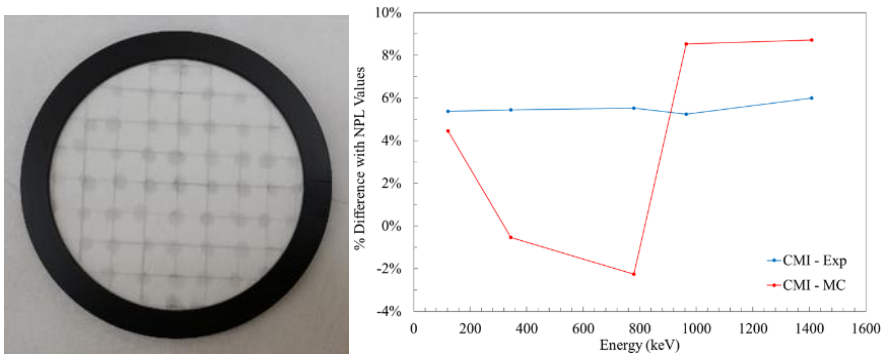

Figure 27: (Left) Filter spiked with ^{152}Eu and (Right) relative differences of FEP DE values, determined experimentally and by Monte Carlo methods by CMI, compared to NPL values determined by measurement of spiked filter. The values exclude correction for true summing.

3.7.4 Conclusion

PTB evaluated the results of two intercomparisons of existing dosimeters used within current dosimetry network systems in European Member States, and novel spectrometers. Information on the traceability, calibration and sensitivity of the dosimetry systems to small dose rate changes has been used by the project to feed into harmonisation procedures.

The implementation of harmonised methods developed in this project will significantly reduce uncertainties in radiological data from typically a factor of 2 or more to a level of 30 % to 40 % for dosimetric data (under reference conditions) and to uncertainties of less than 20 % to 50 % for off-line air-sampling measurements of radioactivity for the most relevant radionuclides released during a nuclear power plant accident.

The traceability of calibrations of activity concentration measurements in air was also improved by this project, especially, by the interlaboratory comparison described in 3.7.3. This is important as traceable calibrations and measurements are a prerequisite for the harmonisation of European early warning networks.

3.8 New and more sophisticated data analysis protocols

A time series of gamma dose rates, measured in the period 1988 to 2015, was analysed. Different periodicities were found and attributed to various physical effects. This knowledge shall be used to reduce uncertainties of dose rate measurements caused by time variations of the natural background radiation.

. A number of different databases were created, which are able to store a large number of spectra provided by early warning network systems. This is especially important for the use of spectro-dosimetry systems and some databases, especially those of IRSN and BfS have been tested in routine operation, by several network operators. There is no final agreement of the best suited database and, hence, several different databases are presently in operation in Europe. In addition, IRSN developed a software tool for data conversion and exchange which will be helpful also for other network operators.

3.8.1 Application of advanced signal processing methods to time series analysis of radiation measurements by early warning systems

Time series analysis of the mean monthly dose rate (measured by the Reuter-Stokes detector in Thessaloniki, northern Greece, from 2001 to 2016) was performed with advanced statistical methods (Fast Fourier Analysis and Zhao Atlas Marks Transform). Fourier analysis reveals several periodicities (periodogram). The periodogram of the absorbed dose rate in air values was compared with the periodogram of the values measured for the same period (2001 to 2016) and in the same location with a NaI(Tl) detector. A quite good agreement was found between the periodograms obtained from the mean monthly dose rate measurements with the Reuter-Stokes high pressure ionization chamber (HPIC) and NaI(Tl) detectors. This is an important result, as the two detectors do not have the same sensitivity to comic radiation. The HPIC detector is sensitive to cosmic radiation (50% of the dose rate in Thessaloniki is due to cosmic radiation) while NaI(Tl) detector is not. With the HPIC detectors the most important periodicity of the dose rate is due to annual cycle. The HPIC detects cosmic radiation and gamma rays mainly originating from soil. Gamma radiation from soil is influenced by different factors, such as precipitation and soil humidity. Many of these factors influencing the gamma radiation are subjected to seasonal variations and therefore the seasonal character is not surprising. The second more pronounced periodicity 46.5 ± 4.7 months is also related to meteorological parameters (air temperature). Apart these two main periodicities, there are also six others (13.3, 17.0, 21.3, 24.4 ,29.5 and 36.6 months) with lower magnitudes which are related to periodicities in solar indices (12.8-13.4 months, 14.5-17.5 months, 22-25 months, 28 months and 31-36 months). Applying advanced statistical analysis to the time series (such as Zhao Atlas Marks Transform) it was found for both detectors (HPIC and NaI(Tl)) that the ≈44-48 months periodicity is very intense during the years 2001 - 2003.

3.8.2 Data evaluation techniques used in the German dose rate monitoring network

The data exchange platform EURDEP collects and provides $\dot{H}^*(10)$ data from all European early warning networks in almost real-time on behalf of the European Commission. Currently, $\dot{H}^*(10)$ data are collected from 30 countries, which have installed almost the same number of different detector types and follow different national policies. The comparability of these data, distributed by EURDEP, is crucial for a meaningful interpretation, not only in the event of a nuclear accident with trans-boundary implications, but also for the correct interpretation of the data under natural background conditions. Furthermore, data assimilation techniques used in decision support systems strongly depend on harmonized data based on both, the physical characteristics of the detectors and on an appropriate description of effects by the measurement site topography.

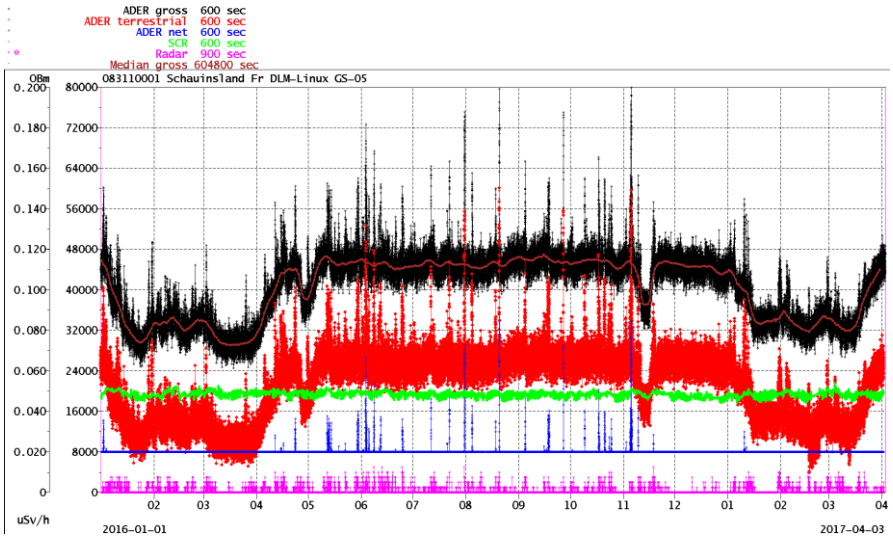

Figure 28: 10 min gross $\dot{H}^*(10)$ (black), median $\dot{H}^*(10)$ calculated over a period of 7 days (brown), SCR (green), terrestrial component (red) and net $\dot{H}^*(10)$ (blue) together with data from the precipitation radar (magenta) obtained at the $\dot{H}^*(10)$ monitoring station at INTERCAL on mount Schauinsland close to Freiburg .

Data harmonization of dose rate data should be based on mechanisms to decompose the ambient dose equivalent rate readings of detectors taking into account the detector's inherent background reading ("self-effect") and the response of the detectors to secondary cosmic radiation. In this way, the terrestrial gamma dose rate can be calculated, which may include, e.g. minor fractions caused by ^{137}Cs from the Chernobyl fall-out and air-borne radioactivity due to radon progeny and which is influenced by soil moisture and possible snow coverage in winter.

As an example of new and more sophisticated data analysis protocols, several recalculated curves of environmental dose rate data are shown in Figure 28. The calculation of net values helps to ease the implementation of automatic systems, which can distinguish rain events from artificial radiological events.

The operation of a network following the design rules and procedures to retrieve harmonized data requires special efforts. Needed steps to obtain the terrestrial dose rate from the raw readings of a detector were published [20]. The calculation of the net $\dot{H}^*(10)$ from all stations in EURDEP may be a first step in the harmonization process. When using this approach, only enhanced net $\dot{H}^*(10)$ values above the statistical fluctuations become visible. In addition, the proposed site characterization procedure should be used. In case of an accident, maps of net $\dot{H}^*(10)$ calculated from EURDEP data would help to identify affected area in Europe.

3.8.3 Conclusion

A time series of gamma dose rates, originally measured in the period 1988 to 2015 on the premises of the University of Thessaloniki in Greece (AUTH), and the different time variations found were attributed by a Researcher Excellence Grant REG(AUTH)) and the project to a variety of physical effects such as solar activity and soil humidity. This knowledge can be used by operators of dosimetric early warning networks to reduce uncertainties of dose rate measurements caused by such time variations of the natural background radiation.

In addition, a number of different databases were created which are able to store a large number of spectra provided by early warning network systems. This is especially important for the use of spectro-dosimetry systems and these databases have been tested in routine operation by several network operators, including BfS in Germany and IRSN in France. There is no final agreement of the best suited database and, hence, several different databases are presently in operation in Europe.

3.9 Underground low-dose calibration facility

The installation of a second low dose rate gamma-ray underground calibration facility in Europe will considerably increase the capacity for the investigation and calibration of dosimetry systems at low dose rates. The idea was to calibrate dosemeters in a collimated photon beam at low dose rates, as being typical in the natural environment. The infrastructure of a new calibration facility at IFIN-HH's underground laboratory (Figure 29), at a depth of 208 m beneath the surface, in the salt mine of Slanic Prahova has been established to allow the calibration of dose rate meters at very low ambient dose equivalent rates in the range from 10 to 100 nSv/h (comparable to dose rates in the natural environment), traceable to CEA's primary dose rate standards. This collimated field underground calibration facility was installed in the salt mine of Slanic Prahova, Romania by IFIN-HH. The low activity concentration of the salt results in a very low background radiation (< 2nSv/h). The shielded collimator for the irradiator has been constructed by the Canberra Packard company.

Figure 29: IFIN-HH's low dose rate underground calibration facility.

In order to characterize the new facility, a first set of measurements has been performed using a radioactive source of Cs-137. During these measurements special attention has been also paid to the determination of the natural radiation background. The background measurements have been done before installing the Cs-137 source in the irradiator. The average measured value of the natural radiation background at the irradiation facilty was $\dot{H}^*(10)_{back}$ = 1,9 nSv/h. This value includes the inherent background of the reference instrument of (0,9 ± 0,9) nSv/h as given in the calibration certificate of PTB for this AUTOMESS instrument.

3.9.1 Radioactive sources for IFIN-HH's new low dose rate underground calibration facility

Different radioactive sources, produced by CEA and CMI, exist for the new IFIN-HH underground facility (Am-241, Co-57, Co-60, Cs-137 and Ra-226). CEA and CMI have characterised the radioactive sources produced for IFIN HH's underground facility by determining the activity and geometry as well as the gamma-ray spectra of the sources.

According to ISO 4037-1, the beta radiation component of the sources must be eliminated. A 5 mm plastic screen with a density 1.35 g/cm^3 (corresponding area mass of 0.67 g/cm^2, thus higher than the value of 0.5 g/cm^2 suggested by ISO 4037-1 were thus placed in front of the sources in the irradiator. Moreover, the spectra of the sources were also recorded by gamma spectrometry with a 0.5 mm stainless steel shield. The emission spectra for each source with both plastic and stainless-steel shields have been characterized by gamma spectrometry.

In addition, the dependence of the ambient dose equivalent rate on the distance r between source and detector has been investigated experimentally. The results show that the $1/r^2$ – law is very well fulfilled.

3.9.2 Secondary standard calibrated against CEA's standard in order to provide traceability for IFIN-HH's new low dose rate calibration facility

An *automess* scintillation dosemeter of the type AD 6150 AD b/H has been selected as secondary standard and has been characterized by CEA in its Cs-137 and Co-60 reference beams. , The inherent background of the instrument was determined at PTB's underground laboratory UDO II.

Figure 14 shows a picture of the AUTOMESS device being irradiated in one of CEA's reference beams and the calibration factors obtained as a function of the ambient dose equivalent rates.

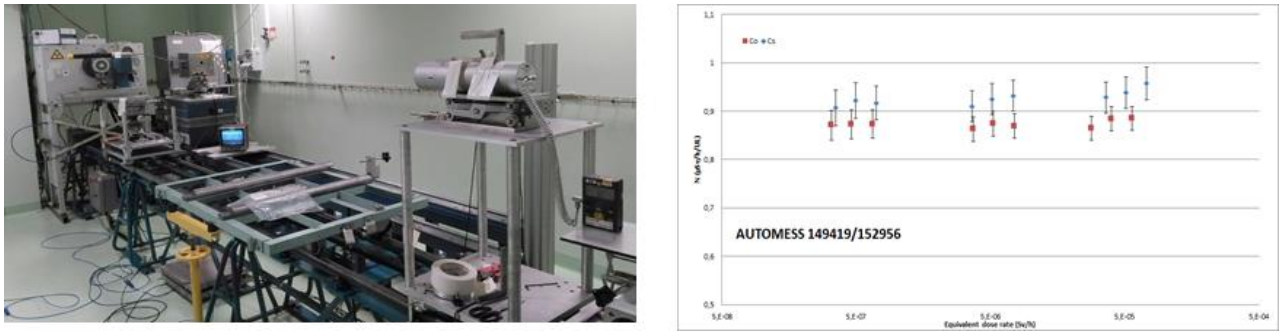

Figure 30: CEA's calibration facility (left) and measured results (right).

In order to study the response function of the secondary standard device, CEA produced three new reference beams in the X-ray range (mean energies 60, 109 and 149 keV) by lowering the dose rates in order to make them compatible with the AUTOMESS device operating conditions (upper limit of recommended range of the AD 6150 AD b/H is 100 µSv/h). The ambient dose equivalent rates achieved at CEA X-ray reference beam are in the range of 30 µSv/h to 50 µSv/h, i.e. the last decade of the dosemeters measuring range, where a slight non-linearity in the order of 10% exists (see Figure 30, measured results).

3.9.3 Conclusion

A new low dose rate gamma-ray underground calibration facility was developed by the project. This facility is only the second in Europe, and considerably increases Europe's capacity for the calibration of dosimetry systems at low dose rates. This new calibration facility is at IFIN-HH's underground laboratory in the salt mine of Slanic Prahova, where the first measurements of dosimeters in a collimated beam at dose rates typical of that in the natural environment were performed.

4 Actual and potential impact

4.1 Metrology achievements

Within this project, standardised measurement and calibration methods were developed and European radiological early warning networks harmonisation improved in:

- dose rate measurement
- airborne radioactivity measurement
- traceability of the measurements
- harmonisation of the European practices

Harmonisation of the measured dose rate data from monitoring systems installed in European early warning network stations was improved to ensure comparability the dose rate data from different European countries reported to a central data base operated by the European Commission. The comparability of the data is crucial

for their meaningful interpretation, not only in the event of a nuclear accident with transboundary implications, but also for the correct interpretation of the data measured under conditions which prevail in the natural environment. To go beyond the state of the art, new instruments based on compact spectrometers were developed and measurement and traceable calibration methods improved, and interlaboratory comparisons organized with participation of project partners and other stakeholders and end-users. [25]

The best practice for the calibration and operation of airborne radioactive contamination detection instrumentation used by nuclear facilities and incident first responders to detect airborne releases in the near field and for instrumentation used by national monitoring networks to detect remote radioactive releases using high volume air samplers was developed. New innovative instrumentation and improvements to spectral analysis techniques was developed enabling the reduction of detection limits leading to the early identification of rising activity levels above ambient background. Studies were undertaken to improve quantification of the effect of radon and other background interferences on instruments that utilise novel detectors. Results were disseminated via peer-reviewed publications, conference presentations and by e-learning modules which enable access to best practice information at locations across Europe via the Internet. Three measuring systems including new measurement and calibration procedures were developed and minimum detectable activities reduced.

The traceability of the measurements obtained by dose rate and air sampling systems installed in early warning network stations were studied, because of the need to further harmonise the European practices in the field of dosimetry and monitoring of radionuclides. Measuring capabilities were enhanced in the field of spectrometry by developing new reference materials, which can be used to test and characterise air sampling and monitoring systems. Dosimetry as well as air sampling or monitoring systems, which are installed in early warning network stations were examined and compared by performing comparison exercises. Conclusions were drawn and recommendations derived to harmonise European monitoring systems so that measured data from different countries are directly comparable.

4.2 Dissemination activities

4.2.1 Scientific publications

Twenty-four high impact publications were published or submitted to key journals, which incorporate the significant scientific outputs of the project (see Chapter 6). A few other publications (not listed in Chapter 6) were and will be submitted for publication in the near future.

4.2.2 Conferences and relevant fora

The work carried out in the project has reached the wider scientific audience at scientific conferences, such as International Conference on Environmental Radioactivity, Global Conference on Radiation Topics (ConRad), ENVIRA 2015, NRC9 2016, NENE 2016, 11th Symposium CRPA, ICRM-LLRMT 2016 and ICRM 2017, EURADOS working groups, EURDEP and AIRDOS meetings. In total, 35 invited lectures, oral presentations and poster presentations were given by the partners during the lifetime of the project. Good feedback was received to all these contributions, which led to discussions and comments. A list of the conference contributions created in the framework of the project can be found in the project public webpage.

Presentations of the outcomes of the project were also given at a number of key fora, in particular at CCRI(II), EURAMET and ICRM working groups' meetings. These activities influenced the future strategy and ensured an on-going impact of the project in the future.

4.2.3 Standards

The project activities and research results were disseminated to following standardisation bodies:

- CENELC/TC45B 'Radiation protection instrumentation'.
- IEC/TC45 'Nuclear instrumentation'/SC45B 'Radiation protection instrumentation'.
- EURAMET TC-IR 'Ionising radiation'.

Within the project, in total 7 particular project activities of JRP, the partners contributed to regulatory bodies, standards organisations, and metrological committees.

4.2.4 Workshops

In order to disseminate the results of MetroERM to stakeholders and to receive input from them on their special needs, MetroERM has funded a stakeholder committee and organized two stakeholder workshops.

The main purpose of the workshops was to inform stakeholders and end-users about the project results and to obtain feedback concerning the joint research of the ten participating European metrology institutes, two radiation protection bodies and two universities.

The first project workshop for stakeholders and end-users was organized and held in Varese (Italy) within the 20nd EURDEP meeting, in March, 2015. The second workshop was organized and held at European JRC in Ispra (Italy), in April 2017. The project research results were presented and discussed and a new project 'Metrology for mobile detection of ionising radiation following a nuclear or radiological incident' was announced. Within both workshops, the following topics were presented and discussed:

- harmonisation of the measured dose rate data from monitoring systems installed in European early warning network stations
- best practice for the calibration and operation of airborne radioactive contamination detection instrumentation used by nuclear facilities and incident first responders to detect airborne releases in the near field and for instrumentation used by national monitoring networks to detect remote radioactive releases using high volume air samplers
- traceability of the measurements obtained by dose rate and air sampling systems installed in early warning network stations, further harmonisation of the European practices in the field of dosimetry and monitoring of radionuclides
- new joint research project 'Metrology for mobile detection of ionising radiation following a nuclear or radiological incident'

At the workshops, participants – stakeholders and end-users – showed very positive interest in the outputs of the project, emphasizing in particular the role of the metrology institutes in the development of metrological standards needed for the comparability of the field measurement results.

4.3 Examples of early impact

Standards and regulation:

The project activities and research results were regularly presented at the CENELC/TC45B 'Radiation protection instrumentation' meetings. Input to a draft documentary standard was realized within IEC/TC45 'Nuclear instrumentation'/SC45B 'Radiation protection instrumentation'/Transportable, mobile or installed equipment to measure photon radiation for environmental monitoring'. The project activities and research results were regularly presented at EURAMET TC-IR 'Ionising radiation' meetings. The project activities and research results were also presented to national nuclear regulators and radiation protection bodies.

End-user uptake:

Czech company NUVIA a.s. is going to install, operate and test the developed modular portable system for airborne radioactivity monitoring, based on mechanically cooled HPGe detector and special low activity concrete shielding. The testing will include validation of the new method for subtraction of natural radionuclides contribution from gamma-ray spectra (so called 'software shielding'). After this about two-year operation and testing, serial production of the system should be realised.

Developed transportable remote controlled compact aerosol sampling device with centrally orientated spectrometric detector for continuous sensitive on-line airborne radioactive particulate monitoring in field stations will be used in the Slovenian NPP Krško. Also, the Belgian research centre SCK.CEN dealing with peaceful applications of radioactivity showed interest to use this system on their site.

Developed continuous on-line particulate airborne monitoring system based on mechanically-cooled HPGe detector will be installed at the Spanish metrology institute reference station ESMERALDA as a part of Spanish early warning network.

European Joint Research Centre organized within the project the 2016 ENV57/MetroERM measurement comparison on simulated airborne particulates: Cs-137, Cs-134 and I-131 in air filters, with dozens of participating laboratories.

Scientific uptake and impact:

The project contributed with 45 presentations at meetings, conferences and workshops and with 7 presentations or contributions to standards, regulatory bodies and working groups. 18 scientific articles were published or submitted to peer-reviewed scientific journals. 5 trainings for project partners and stakeholders and 2 workshops for stakeholders were organised.

4.4 Potential impact

Within the project, novel and improved instrumentation and new measurement techniques and analysis methods were developed for field stations of the radiological early warning networks in Europe. The metrological foundation and particularly, the traceability of measurements carried out at these stations were improved significantly, thus addressing the harmonisation of the European early warning networks.

The project outputs contribute to the improvement of the quality of dose rate and air borne radioactivity data derived from the measurements of the 5000 radiological early networks stations in Europe and together with the new and more sophisticated data analysis methods developed in the project contribute to a faster and more coordinated response of European authorities in the case of a nuclear emergency. The quick and reliable assessment of radioactive contamination and dose rate levels is of key importance for the effectiveness and optimisation of countermeasures for the protection of the public health and the environment. The project outputs contribute to the recommendations of the European Commission to EU Member States, based on data from the national early warning networks. The quick and reliable assessment of radioactive contamination and dose rate levels is of key importance for the effectiveness and optimisation of countermeasures for the protection of the public health and the environment.

Specifically:

- More precise area dose rate and air borne radioactivity data are derived from the measurements of the almost 5000 radiological early warning network stations in Europe. Quality systems for calibrations traceable to primary standards were analysed and advice provided on how to harmonise the associated procedures.
- New measurement techniques and traceable measurement procedures are available aimed at the harmonisation of calibration procedures and the implementation of good practice guidance. The implementation of these significantly reduces uncertainties from a factor of 2 or more to a level of 30 % to 40 % for dosimetric data (at least for dose rates between 100 nSv/h and 200 nSv/h and under reference conditions) and to uncertainties of less than 20 % for gamma emitting nuclides, and 50 % for pure beta- and alpha-emitters for off-line air-sampling measurements of radioactivity for the most relevant radionuclides released during a nuclear power plant accident.
- Basic metrological information (performance parameters) is provided for dosimetry, spectrometry and air sampler systems for metrologically sound measurements of dose rates, contamination levels on ground and air contamination levels in real-time. This information is a prerequisite for any competent governmental decision and appropriate countermeasures in the case of a radiological emergency with transboundary implications.
- Metrological developments for the improvement of the European early warning networks in the field of airborne radioactive aerosol and dose rate measurements allow an early indication of a nuclear threat, whether in the event of a nuclear accident (such as the nuclear power plant accidents of Chernobyl and Fukushima) or caused by other radiological incidents.
- Best practice derived from the comparison exercises and the evaluation of differing calibration procedures is disseminated via e-learning material made permanently available on the internet utilising web-pages already established for the user communities.

Environmental impact:

An early indication and better determination of the affected areas allow the identification of radionuclides and contamination levels in real-time, and harmonised, coordinated and more adequate and efficient countermeasures to protect the affected population. The harmonised area dose rate data also allow investigations into parameters affecting climate change, e.g. soil moisture in real-time, using Europe-wide dose rate mappings of European Radiological Data Exchange Platform (EURDEP).

Social impact:

Reliable radiological data from routine measurements are required in order to achieve credibility and acceptance by the public of reported data, particularly in the event of a nuclear emergency. Harmonisation between national early warning networks is therefore essential to “eliminate political borders in contamination patterns”, especially close to national borders. In an emergency, reliable and traceable real-time information on dose rate and ground contamination levels as well as on airborne radioactivity concentrations significantly supports adequate and harmonised governmental decisions and countermeasures on a European scale.

Financial impact:

Harmonisation of existing radiological early warning networks and the implementation of new measurement techniques for radioactive aerosol measurements improve the protection of the general population against the dangers of ionising radiation, in particular during the early phase of an accident, and minimise the follow-up costs. Greater accuracy in the determination of the extent of land contamination help to reduce the area designated for exclusion and evacuation zones. The early estimation of the contamination of agricultural products leads to swifter decisions on whether products need to be banned from the market. Follow-up costs after the significant nuclear accident (like Chernobyl) can be estimated to tens of billions of Euros.

5 Website address and contact details

A project public website has been created: www.earlywarning-emrp.eu. A partners’ share point has also been created, in order to give the possibility to all the partners to share work documents and deliverables.

The contact person for general questions about the project is Stefan Neumaier, PTB

(stefan.neumaier@ptb.de).

The contact person for dose rate monitoring networks is Harald Dombrowski, PTB

(harald.dombrowski@ptb.de).

The contact person for airborne radioactivity networks is Steven Bell, NPL (steven.bell@npl.co.uk).

The contact person for the traceability of IFIN-HH’s underground calibration facility is Valentin Blideanu, CEA

(valentin.blideanu@cea.fr).

The contact person for fast radiochemical methods is Daniel Zapata, PTB (daniel.zapata@ptb.de).

The contact person for the public website and the exploitation and dissemination of the projects results as well as the CMI airborne radioactivity system is Petr Kovar, CMI (pkovar@cmi.cz).

The contact persons for the CIEMAT and IJS airborne radioactivity monitoring systems are Nestor Armando Cornejo, Ciemat (nestorarmando.cornejo@ciemat.es) and Denis Glavic-Cindro, IJS (denis.cindro@ijs.si), respectively.

The contact persons for the JRC partners involved are Marc de Cort, JRC-Ispra (marc.de-cort@ec.europa.eu) and Mikael Hult, JRC-Geel (mikael.hult@ec.europa.eu).

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