ABSTRACT

The main aim of the European metrological research project Metrology for radiological early warning networks in Europe (MetroERM) is to improve the metrological foundation of measurements of fundamental radiological quantities such as ambient dose equivalent rate, radioactivity concentrations in air and ground contamination levels in real-time, as well as 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.

In the frame of this project a compact portable aerosol sampling and measurement device was developed at Jožef Stefan Institute. The system incorporates a CeBr₃ scintillation detector positioned centrally within a concertinaed filter assembly and an improved high flow
rate air pump. It provides continuous on-line low level airborne radioactive particulate monitoring for field station use connected directly via 3G network communication to diagnostic center. The calibration of the device was performed at National Physical Laboratory (NPL) with filters spiked with a certified mixed nuclide solution. Additionally first tests were performed in an environment with an elevated radon concentration.

1 INTRODUCTION

In a major radiological emergency early and reliable knowledge of radioactivity concentrations in air and the subsequent assessment of contamination levels of farmland and of dose rate levels in urban areas are of key importance in order to organise appropriate countermeasures for the protection of the general public against dangers arising both from direct external radiation and from intake of radioactivity from foodstuff and from air. Fast and reliable information is an essential prerequisite for governmental decisions and has a considerable financial impact.

The nuclear power plant accident at Fukushima Daiichi clearly confirmed the need for exchanging radiological information in real-time at an international level, even for remote accidents. During a radiological emergency with trans-boundary implications in Europe, the European Commission will issue recommendations to EU Member States based on data from national early warning networks. This 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, coordinated with data from international radiological networks, are a prerequisite for adequate environmental radiation monitoring in Europe.

With the aim to deliver metrologically sound measurements of fundamental radiological quantities such as ambient dose equivalent rate, radioactivity concentrations in air and ground contamination levels in real-time, the 3-year EMRP joint research project Metrology for radiological early warning networks in Europe (MetroERM) [1] was launched in 2014. Within this project collaborate European metrology and research institutes, JRC at Ispra which is responsible for the EURDEP database, as well stakeholders and manufacturers of radiological monitoring detector systems. This establishes the basis for unique possibility to comprehensively address the harmonisation of the radiological early warning networks in Europe - the largest and most comprehensive environmental radiation monitoring system worldwide.

The main achievements, planned within this project, are consistent data collection and evaluation as well as harmonization of reported values on both dose rate and airborne radioactivity concentrations so that data related to the same trans-boundary event measured by different networks using different detectors are directly comparable and consecutively reliable conclusions that could be drawn by the responsible authorities. In addition, within this project the development of new measurement techniques based on spectrometry systems with novel detection materials is in progress with the aim to allow both the calculation of dose rates and the calculation of contamination levels including nuclide-specific information in real time.

Another important aim of MetroERM project is to improve the capacity of the early warning networks by the development of new methods and systems for rapid and precise measurement radioactivity-in-air at low concentrations. In this way the global early warning data would be efficiently supplemented with accurate information on airborne radionuclide content including nuclide-specific information. New in-situ spectroscopy systems for airborne radioactive particulate monitoring have been developed on the basis of comprehensive
investigations of detector features and of spectral evaluation and deconvolution methods for new and improved measurement systems based on spectrometric detectors like HPGe, CdZnTe, La(Ce)Br₃, CeBr₃ and Cs(Tl)I [2]. These systems provide real time information of radioactive aerosol concentration levels and will become the core instrumentation of the next generation of environmental radiation monitoring networks in Europe.

2 SCIENTIFIC AND TECHNICAL OBJECTIVES OF THE METROERM PROJECT

All European countries operate airborne radioactivity and dose rate early warning networks. There are about 5000 dose rate monitoring stations operational in Europe, which provide hourly data transmission to the European Radiological Data Exchange Platform (EURDEP) operated by the European Commission. However, only approximately 250 of these stations are capable of on-line particulate and/or gaseous airborne monitoring. In most cases, the collected data require further analysis as neither appropriate calibration nor corrections for the variety of detector types have been performed. Moreover, due to the simple detector designs, like Geiger Muller counters, which are typical used in dose rate networks and their pronounced energy and angular dependent response to gamma radiation, as well as their calibration at single photon energy, it is unreliable to derive correct dose rate data. As a consequence, the measured values may differ by a factor of 2 or more even under the same measuring conditions, and hence during a trans-boundary nuclear incident contradictory decisions may be taken in different countries.

As the quantity of data submitted to the EURDEP database has increased, network operators and metrology institutes have become more aware of the impact of the current high uncertainties in the area dose rate and airborne radioactivity concentration measurement data. Action on a European scale is required to address the underlying measurement problems and to resolve them. The aim of this project is to improve the metrological foundation of measurements, including 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. The specific scientific and technological objectives are:

- To develop novel and improved instrumentation for field station use to enable both the measurement of dose rates and the collection of nuclide-specific information.

- To undertake comprehensive scientific investigations of detector features and of spectral evaluation and deconvolution methods for new and improved measurement systems based on “high-resolution” non-cryogenic spectrometric detectors, e.g. LaBr₃, CeBr₃, SrI₂ and CdZnTe. In parallel, to develop improved instrumentation for the field of airborne radioactive particulate monitoring.

- To validate common metrological procedures and to implement traceable calibrations of detector systems used to supply data to central databases, especially EURDEP.

- 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.

- To validate new detection principles such as the use of the new spectrometry systems for the calculation of dose rates and contamination levels from in-situ spectra by Monte Carlo simulations and bench mark experiments.
– To undertake enhanced on-site evaluation of the diverse environmental and radiological conditions and measurement techniques used at monitoring stations (provision of background information on site conditions and scientific development of appropriate correction methods).

– 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.

– To develop 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 and dosimetry measurements at field stations.

– To develop new and more sophisticated data analysis protocols to enable rapid information dissemination.

The MetroERM project is subdivided into five work packages (WP):

– WP1 develops novel and improved instrumentation for spectrographic dose rate and dose rate measurements for field station use, validated by benchmark experiments, long-term measurements and Monte Carlo simulations.

– WP2 improves existing and develops new instrumentation for the determination of airborne radioactivity concentration and develops improved techniques for the validation of gamma-spectrometry data.

– WP3 addresses the "traceability" of dose rate and airborne radioactivity measurements, by the quantification of improved metrological procedures and an adequate quality management system. Furthermore, WP3 explicitly supports the process of harmonisation of radiological data from early warning networks in Europe by systematic investigations, comparison exercises and the publication of recommendations.

– WP4 aims to maximise the impact of the JRP by dissemination of information and developments from the JRP to national network operators via e-learning and web-based materials, through workshops aligned to stakeholders’ needs and presentations at international conferences, and publications in peer-reviewed journals.

– WP5 describes the management of the JRP.

By the end of the JRP, faster and more precise measuring instruments and methods will be available, for both dosimetry and airborne radioactivity concentration measurement. The current sources of discrepancy will be clearly identified. Solutions to resolve them will be proposed and disseminated to stakeholders and to end users via web-based materials, at conferences and in journals. The data transmitted to the EURDEP database will be more consistent and methods will be developed to correct the identified current inconsistencies in input data. This will lead to reliable information on validated area dose rate and activity values being available to inform decisions on the radiological situation across Europe during a nuclear incident.
3 THE COMPACT RADIOACTIVE AIRBORNE PARTICULATE MONITORING INSTRUMENT DEVELOPED AT JSI

At Jožef Stefan Institute (JSI) a novel compact portable aerosol sampling device was developed within the work package WP2. The device incorporates a CeBr$_3$ scintillation detector positioned centrally within a concertinaed filter assembly and an improved high flow rate air pump. The system provides remote and continuous on-line low level airborne radioactive particulate monitoring from field station to decision makers server via 3G network communication. The entire device is assembled into a heavy-duty portable Peli Case 1740 with exterior dimensions of 37 cm (height without cover), length of 114 cm and width of 50 cm. A schematic render of the entire system assembly is shown on Fig. 1. The system has been divided into three subsystems based on their functionality: air management, detection and processing subsystem.

![System Assembly Diagram](image)

**Figure 1:** Air pump system assembly (A):
- a) concertina aerosol filter
- b) CeBr$_3$ detector with integrated bias voltage supply
- c) flow meter
- d) air pump motor and turbine (brushless blower)
- e) microcontroller unit with TFT touch screen and I/O controls
- f) preamplifier and digital pulse processing unit
- g) 230 V AC power IEC connector and ON/OFF switch with LED indicator
- h) 2 USB connectors for firmware upgrades and serial communication with a PC.

Image (B) illustrates the filter assembly, multiple arrows indicate entry of air through the filter and the single arrow shows airflow output into the flow meter. Image (C) presents a top view of the entire system in a rugged Peli Case.
3.1 Air management subsystems

The air unit consists of a concertinaed aerosol filter assembly, a flow meter and a high-performance air pump. The pump provides a stable flow rate up to 200 m³/h. The concertina aerosol filter (Fig. 1a) has a tubular shape, so that the air flows through the filter and the deposited aerosols on the filter are constantly being measured by the CeBr₃ detector which is fitted inside the filter (Fig. 1b). The air flows through a flow meter (Fig. 1c) and exits the system through the air pump (Figure 1d). Filter cartridge consists of a metallic housing where 0.87 m² of fiberglass H13 class filter paper folded in an accordion form is placed. At air-flow rates used, the collection efficiency of 99.99 % for particles with sizes above 0.3 µm. The air output from the assembly continues through a Testo 6444 flow meter, mounted to the filter assembly. The flow meter records volume flow according to the calorimetric principle, keeping the measurement results independent of the process pressure and does not cause a permanent pressure drop. It encodes real time data on the instant and integrated total air flow into a digital pulse stream, suitable for readout by the microcontroller unit.

3.2 Detection subsystems

Since the device should be portable, special care was taken in choosing a suitable size for the detector. It has to fit in the concertina aerosol filter without significant impact on the airflow rate. Due to their best cost/benefit ratio we decided to use a scintillation detector. Two fairly new scintillation materials, lanthanum bromide (LaBr₃:Ce) and cerium bromide (CeBr₃) were considered alongside the classic, cesium iodide (CsI:Tl). Since the device is made for an early warning system, it needs to be as sensitive as possible for low activities, therefore a low inherent background is required. On this basis the main decision parameters for detector choice were energy resolution and intrinsic radioactivity. With these considerations in mind we selected a 1 inch (2,54 cm) CeBr₃ detector with emphasis on purity of the crystal itself [3, 4, 5]. The detector has a FWHM energy resolution of ~4 % at 662 keV. The bias voltage generator is already incorporated into the detector housing, additionally a CR-112 charge sensitive preamplifier module by Cremat Inc. is used to amplify and shape pulses from the detector. The signals from the detector are digitized and processed by a Digital Signal Processing unit (DSP unit), developed in-house at JSI (Fig. 1f), based on a field programmable gate array (FPGA) and a 14-bit analog to digital converter (ADC). The pulses are collected in 4096 channel-wide spectra (4k), sufficient to match the detector energy resolution. The detector communicates with the base unit over a serial link.

3.3 Processing subsystems

All measured parameters and system settings are controlled by a microcontroller unit with a color touch-screen 800x480 TFT display supporting a graphic user interface (GUI) for easy interaction with the system in the field, or via a 3G connection (Fig. 1e). This part of the system is called base unit and it supports connection to an external computer to perform heavier real time computing, make basic decisions, communicate and send data to the user. Since the system is designed for early warning scenarios, the communication between a group of systems in the field and a remote (central) safe working area is needed, therefore a microcontroller-based system with significant processing power is needed to coordinate all functions and perform parts of the data analysis already in the field. These specifications were economically met by a Mikromedia 5 for Tiva development board. The board supports an add-on 3G module for remote operation and data transfer. An integration I/O board was
developed in-house to inter-connect all the subsystems. When the system is switched on, the base unit boots into an idle state and the main menu is shown. From there, the user can start the device, set various parameters and start a measurement. There are two ways to retrieve measured data from the system, by a serial-over-usb connection to a personal computer or by sending a request in an SMS message to receive same data via e-mail. The system also supports local storage, so that all data can be saved locally to be retrieved and analyzed later.

3.4 Instrument testing and calibration

The detection system was initially tested with a Cs-137 point check source with an activity of 56.25 kBq at reference date 18. 4. 1994. The source was positioned 4 cm from the detector on its symmetry axis. The amplified signal from the detector was analysed by our digital pulse processing unit. After 2500s of measurement live time the FWHM of 4.4 % at the 662 keV photo-peak was determined by Gamwin software.

The validation of the compact radioactive aerosol particulate monitoring device was performed at the National Physical Laboratory (NPL) in Teddington, UK. 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 (Am-241, Cd-109, Co-57, Ce-139, Cr-51, Sn-113, Sr-85, Cs-137, Mn-54, Y-88, Zn-65, Co-60 and Y-88). The relative activities of the radionuclides in the mixture was chosen so that the relative intensities of the peaks in the gamma-ray spectrum were comparable. 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 for 3440 seconds without pumping air through the filter. The gamma-ray spectrum collected during this measurement is presented in Fig. 2. Since the exact activities of each radionuclide spiked on the filter are known a priori, this measurement was used for determination of the photo-peak efficiency calibration curve. The photo-peak efficiency was calculated at several energies between 60 keV (Am-241) and 1836 keV (Y-88) from the measured net areas for each photo-peak, the branching ratio for gamma emission and half-lives of the radionuclides. The plot of efficiency curve is shown in Fig. 3.

As is evident from Fig. 3, the total photo-peak efficiency peaks at slightly less than 1 %, consistent with the size of the detector crystal and the average distance from the filter paper. Expecting that the aerosols would be distributed homogenously over the entire filter surface in real measurements with the air pump turned on, we can use this calculated efficiency curve for the calculations of actual airborne activities in the field.

After 25 hours of the measurement of the low activity filter only peaks due to natural radioisotopes were present in the spectrum. On this basis we concluded that the deposited activity on the filter in this case is under the system detection limit.
Figure 2: Energy spectrum of a high activity filter measured for 3440 seconds. Peak at 1580 keV is an artefact due to erroneous setting of upper limit threshold on DSP unit.

Figure 3: Photoabsorption peak efficiency, calculated from the high activity filter data.

3.5 In-field testing of the instrument

One field test of the full function of the aerosol particulate monitoring device was performed at NPL in the basement room of Bushy House where elevated levels of radon and thoron progenies are present. For this measurement we used a fresh filter and the air volume of 5000 m³ was being pumped through the system for approximately 25 hours. In parallel, the total activity of radon was also measured with a passive radon diffusion chamber Canary Radon Meter, recording an average activity of $(450 \pm 50)$ Bq/m³. The measured spectrum
obtained with the aerosol particulate monitoring device is shown in Fig. 4 where spectral lines originating from radon and thoron progeny are clearly seen.

To perform a quick check of our relative efficiency calibration, we compared the calculated activity of Pb-214 as obtained individually from two spectral lines of Pb-214 and got the same value both times. This validated our determination of efficiency curve. If radon progenies were in equilibrium with radon, calculated radon activity from Pb-214 and Bi-214 activities should be the same. This was not the case, so we assume that they were not in equilibrium, most likely due to the sudden air exchange when we entered the chamber during setup.

Figure 4: Energy spectrum as measured at Bushy house basement where radon and thoron progeny are present.

4 CONCLUSIONS

The compact radioactive aerosol particulate monitoring device developed at JSI provides continuous, sensitive, on-line airborne radioactive particulate monitoring for field station use. With the defined energy calibration and photo peak efficiency, one can accurately determine airborne gamma activities.

The main advantage of the system is that the compact radioactive aerosol particulate monitoring device is incorporated in a heavy-duty portable case which is easily transportable to different measurement locations. As the air pump enables high flow rates, also low activity airborne radionuclides can be quickly measured. With prompt and continuous online detection and 3G connectivity it enables human-error free data relay from remote field stations to a centralized system for evaluation of gamma emitting radionuclides in spectra, as well remote control over the unit settings and function. During a nuclear emergency increased frequency of data transmission is possible, which enables remote analysis of plume deposition.

The existing device in its current form only accumulates gamma-ray spectra and sends them to the remote stations and no spectral analyses are performed at the sampling location yet. The further developments and upgrades of the portable air-sampling system are firmware update with a nuclide ID algorithm and software development that enables analysing the data...
and providing estimates on absolute radioisotope concentrations in air at the sampling locations.

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REFERENCES


