Radiation Measurements 146 (2021) 106615

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Investigations into the basic properties of different passive dosimetry systems used in environmental radiation monitoring in the aftermath of a nuclear or radiological event

Željka Knežević [a,*], Marija Majer [a], Zuzanna Baranowska [b], Olivera Ciraj Bjelac [c], Giorgia Iurlaro [d], Nikola Kržanović [c], Francesca Mariotti [d], Marijana Nodilo [a], Stefan Neumaier [e], Katarzyna Wołoszczuk [b], Miloš Živanović [c]

[a] Ruđer Bošković Institute,(RBI), Croatia
[b] Central Laboratory for Radiological Protection (CLOR), Poland
[c] Vinca Institute of Nuclear Sciences (VINS) - National Institute of the Republic of Serbia, University of Belgrade, Serbia
[d] Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA), Italy
[e] Physikalisch-Technische Bundesanstalt (PTB), Germany

ARTICLE INFO

Keywords:
Passive dosimetry systems
Environmental radiation monitoring
Nuclear or radiological event

ABSTRACT

Due to the need for harmonization of passive dosimetry and the requirements of the international standards in the area of environmental monitoring in radiation protection, multiple types of various passive dosimetry systems based on different ionizing radiation detection mechanisms were subjected to extensive performance testing. In the scope of the EMPIR project 16ENV04 Preparedness, the performance of 12 passive dosimetry systems was examined in radiation fields of different photon energies, angles of incidence and ambient dose equivalent rates in order to estimate their performance in the almost omni-directionally and energetically broad radiation field of the natural environment. The use of different detectors, holders, calibrations, measurement procedures and uncertainties, leads to differences in the measured data. Prior investigations and harmonization of passive dosimetry systems are necessary to achieve reliable and comparable dose measurements in Europe.

1. Introduction

In the aftermath of a nuclear or radiological event, accompanied by the release of a large amount of radioactivity, long-term monitoring of large contaminated areas may become necessary in order to determine the external gamma dose to the public and to support governmental decisions on counter measures, like e.g. the proclamation of exclusion zones and the resettlement of its inhabitants. Such decisions may affect thousands of people and the need for environmental radiation monitoring in the affected areas, like around the Chernobyl nuclear power plant, may remain for years or even for decades. Passive dosemeters are small, easy to handle, cheap and robust and they don't need any electrical power supply. Therefore, such systems are well suited for the purpose of long-term monitoring of ionizing radiation in the environment following a nuclear or radiological accident.

Passive dosimetry systems in general are widely used in environmental radiation monitoring (Budzanowski M. et al., 2004; Dombrowski H., 2019; Dombrowski H., Neumaier S., 2012; Duch M. et al., 2017; Duch et al., 2021; Nanto, H. et al., 2011; Ranogajec Komor, M., 2009; Saez-Vergara, J. C., 2000), especially for long-term dosimetry measurements around nuclear installations. The most widely used passive dosemeters for this purpose are thermoluminescence (TL) dosemeters. Other passive dosimetry systems are based on optically stimulated luminescence (OSL) and radio-photoluminescence (RPL) dosemeters. Some of the systems routinely used in environmental radiation monitoring are developed and applied for personal monitoring (Nanto, H. et al., 2011; Ranogajec-Komor, M., 2003; Sarai, 2004). Despite their widespread use in the area of environmental monitoring, there is still a lack of international standardization and harmonization of environmental monitoring by passive dosemeters across Europe. The current IEC 62387 (2020) standard sets out requirements for the properties of the detector systems and enables detailed planning for the type testing procedures. But at present, general guidance and recommendations on the routine measurement procedures, for environmental radiation

* Corresponding author.
E-mail address: zknez@irb.hr (Ž. Knežević).

https://doi.org/10.1016/j.radmeas.2021.106615
Received 11 January 2021; Received in revised form 28 May 2021; Accepted 18 June 2021
Available online 24 June 2021
1350-4487/

monitoring does not exist. There is not much data in the literature either, except for intercomparison exercises (Dombrowski, H., 2019; Dombrowski, H., Neumaier, S. 2012) which are focused on systematic investigations and comparisons of different passive dosimetry systems used by different institutions in environmental monitoring.

A study of the uncertainty budget and detection limits of passive dosimetry systems used for ambient monitoring was performed within Preparedness project, by applying ISO standard 11929 (ISO, 2019) on the 4 dosimetry systems used by CLOR, ENEA, IRB and VINS. The study considered the application in normal as well as in emergency situations. It was found that the only common uncertainty component for all four laboratories was the reader sensitivity factor, and the detection limit depends on the number of parameters included in the uncertainty budget. The harmonization of the methodologies of environmental dose computation is necessary for a correct comparison of the detection limits of different dosimetry systems (Iurlaro, G., 2021).

The objective of this work is to investigate the properties of different passive dosimetry systems used by different European institutions in environmental radiation monitoring, including dose response, energy response, angular response and the response of the passive systems to a natural radiation spectrum. This research has been performed within the European Metrology Programme for Innovation and Research (EMPIR) by the project 16ENV04 “Preparedness” (Neumaier, S. et al., 2019; Preparedness Consortium, 2019, www.euramet.org).

2. Materials and methods

2.1. Dosimetry systems

In the measurement campaign to determine the main characteristics of typical passive dosemeters, the following 9 institutions participated with 12 different systems: Central Laboratory of Radiological Protection (CLOR), Poland; ENEA-Radiation Protection Institute Italy; Institute for Occupational Health (IMI), Croatia; Jožef Stefan Institute (JSI), Slovenia; L.B. Servizi per le Aziende SRl (LBSA) Italy; Politecnico di Milano (POLIMI), Italy; Ruđer Bošković Institute (RBI), Croatia; Universitat Politècnica de Catalunya (UPC), Spain; Vinca Institute of Nuclear Sciences (VINS), Serbia.

Nine out of 12 systems used different types of TLDs and the remaining three were based on RPL, OSL and film dosemeters. The details about the used systems, detector types, holders and readers are summarized in Table 1 and Fig. 1. The passive dosimetry systems for environmental monitoring are usually employed in groups of several individual detectors, allocated in a suitable holder with plastic and metallic filters to provide the adequate filtration for the measured quality as well as to protect the detectors against dirt, liquids or light which is particularly important in environmental radiation monitoring. The angular and energy dependence of the response of a dosemeter may be influenced by its design, especially by an energy compensation filter in the holder.

The dose calculations were performed by each institution according to their own routinely used procedures. Preparation, calibration and readout procedures, as important part of the dose evaluation, were carried out according to these standard procedures. For all results, transport dose corrections were taken into consideration and all the resulting doses are expressed in the quantity ambient dose equivalent ($H^*(10)$) with the corresponding unit Sv (Sievert) or in parts of it (e.g. mSv = milli Sievert).

2.2. Irradiation set-ups

Irradiations of the various passive dosimetry systems were carried out in four Standard Dosimetry Laboratories (SDL) using their respective irradiation facilities (i.e. the reference doses are traceable to primary standards) in order to determine the dosimetric properties of these systems.

2.3. Basic properties of the systems

The response of passive dosimetry systems is studied for various influence factors. In this context, the response (r) of a dosimetry system is defined as the ratio of the dose reading of this system after an exposure (eventually corrected for inherent background, fading and other influence factors) and the reference dose value (“true” dose value) for the irradiation scenario, i.e. r = measured dose/reference dose. For an ideal measurement r = 1. In the following, such absolute response values are called “ absolute response”. In addition, it may be advantages to normalize the “response function” (if the dependency of the response

Table 1
Summary of passive dosimetry systems used in the study.[a]

Participant	Dosemeter type	Detector/commercial name	Detector holder	Reader type	[a]Relative uncertainty in dose estimation
A	TLD	LiF:Mg,Ti/TLD-100	ABS (Cycolac) holder + sealed waterproof plastic bag	Harshaw 5500	10–33% (k = 2), $H^*(10)$
B	TLD	CaF_2:Mn/(TLD-IJS-05), LiF:Mg,Cu,P/(MCP-n) Al_2O_3:C/TLD-500	packed in rubber lined polyethylene bags and inserted in plastic box holder sealed in waterproof plastic bag	Toledo Vinten	22% (k = 2), $H^*(10)$
C	TLD	LiF:Mg,Cu,P/(MCP-N, RADCARD)	4 elements card (open window, plastic filter, 2 aluminum filters)	RADOS RE 2000	17% (k = 2), $H^*(10)$
D	TLD	TLD-2000C. Conqueror Electronics Technology	PVC Holder packed in a sealed waterproof plastic bag	Harshaw 5500	16% (k = 2), $H^*(10)$
E	TLD	LiF:Mg,Cu,P/TLD-700H (07H7H0/TLDCARD-27P)	2 element card holder; PTFE foils, and mounted on an aluminium plate.	Harshaw 6600 PLUS	24% (k = 2), $H^*(10)$
F	TLD	CaF_2:Mn/(TLD-IJS-05), LiF:Mg,Cu,P/(MCP-n)	packed into the plastic bag	TLD-MR-200 C, in-house reader	around 22% (k = 2), $H^*(10)$
G	RPL	Ag activated phosphate glas (type FD-7)	holder type SC-1; holder with Sn filter sealed in waterproof plastic bag	FGD-202E	19% (k = 2), $H^*(10)$
H	OSL	Al_2O_3:C/InLight Landauer	4 elements card (open window, aluminum, copper and plastic filters)	MicroStar Landauer	12–52% (k = 2), $H^*(10)$
I	TLD	LiF:Mg,Cu,P/(GR-200A)	2 elements card assembled between two ABS cards	Harshaw 6600 PLUS	25% (k = 2) $H^*(10)$
J	FILM Symmetric	AGFA PMF 22MUO	multy filter system (Air, 0.05 mm Cu–0.3 mm Cu-1.2 mm Cu–0.8 mm Pb)	X-Rite 361T Densitometer	50% (k = 2), $H^*(10)$
K	TLD	LiF:Mg,Cu,P/(GR-200A)	2 elements card assembled between two ABS cards	RADOS RE2000	50% (k = 2), $H^*(10)$
L	TLD	$CaSO_4$	Panasonic, 3 element card, filtration 0.7 mm Pb sealed in waterproof plastic wrap	Panasonic UD-716	25% (k = 2), H^* (10)

[a] Reported uncertainties are estimated for different choice of parameters in the uncertainty budget used by respective laboratory and calculated according their own procedure. The stated uncertainty range varies with the dose: low dose, greater uncertainty; high dose, lower uncertainty.

Fig. 1. Different types of dosimetry systems used.

is studied for a given influence factor, like e.g. energy of the photons, in the following also called "energy response"), i.e. for a special reference value of the influence factor, the corresponding response value is set to one (r = 1). In case of the energy response, the normalization to S–Cs irradiation (i.e. 662 keV photon energy) is frequently used; hence r (662 keV) = 1. Response values normalized in such a way are in the following called "relative response" (or normalized response function).

The following dependencies of the response of the passive dosemeters described in section 2.1 are studied:

2.3.1. Dose dependence

A dose dependence test was carried out at the SSDL of the Ruđer Bošković Institute (Vekić, B. et al., 2006). Ten different values of air kerma free in air in the range of 0.15 mGy–1 Gy were selected and 5 dosemeters per system and per dose were irradiated. The irradiations were performed by employing two S–Co sources with different activities (a radiotherapy S–Co source with dose rates between 88.53 mGy/min to 102.325 mGy/min and a radiation protection S–Co source with dose rates between 2.168 μGy/min and 2.506 μGy/s) in order to cover a wide range of doses. The reference dose rates were verified by secondary standard (ionization chambers types PTW farmer 30013 and PTW LS-01 32002) in terms of air kerma free in air. The irradiation doses were converted into $H^*(10)$ by using the conversion coefficient stated in ISO 4037-3 (ISO, 1999) and the obtained values were: 0.18 mSv, 0.36 mSv,

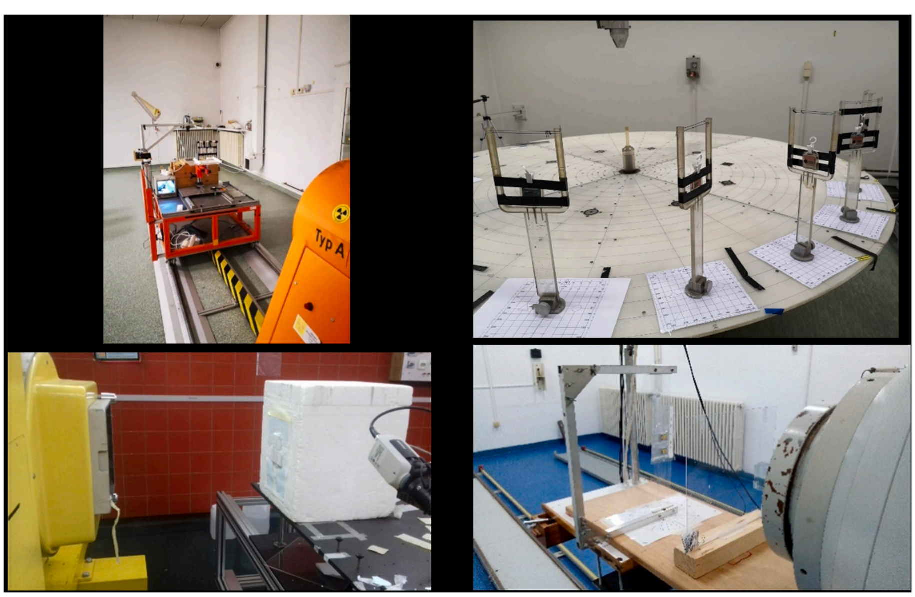

Fig. 2. Gamma stand, S–Co and S–Cs sources (upper left) and Ra-226 stand (upper right) at CLOR, S–Co at RBI-SSDL (lower left), and S–Co (lower right) at VINS- SSDL.

0.6 mSv, 1.2 mSv, 6 mSv, 24 mSv, 60 mSv, 0.12 Sv, 0.6 Sv and 1.2 Sv. The dosemeters were irradiated free in air at 1 m distance from the source (Fig. 2). The uncertainties of the dose in reference irradiations with S–Co sources were 4.3%, (for a radiotherapy S–Co source) and 4.6% (k = 2) (for radiation protection S–Co source).

The dose response of different passive dosimetry systems was assessed as the absolute response obtained by dividing the ambient dose equivalent mean value for 5 irradiated dosemeters by the reference value.

2.3.2. Angular dependence

Angular dependence tests were carried out at the SSDL of VINS, Serbia. Reference values were established according to ISO 4037, using a secondary standard calibrated in terms of air kerma free-in-air. Conversion to *H**(10) was performed by using conversion coefficient from ISO 4037 (ISO, 1999). A total of 12 dosemeter types were irradiated in S–Cs radiation quality. Irradiations were performed at 5 different angles: 0°, 30°, 60°, 90° and 180°. Reference orientation and reference point were defined for each system by the laboratory that provided the dosemeters, in order to test the performance of the system as used by the service. The irradiation distance (120 cm) was adequate to ensure that the whole dosemeters were irradiated. Two dosemeters were irradiated at the same time, with 2–4 irradiations per angle (Fig. 2). The delivered dose was 2 mSv, except for (E) dosemeters, which received 5 mSv. The difference is due to the fact that the (E) dosemeters were irradiated by a stronger source before this irradiator showed a malfunction. The uncertainty of the delivered dose was between 4.8% and 5.0% (k = 2). The angular response was assessed as relative response, calculated as the ratio of the dosemeters reading for different angles of incidence and the reading for 0° angulation. It should be noted that some of the dosemeters were not symmetrical, so choosing a different axis or direction of rotation would produce different results. However, the test included irradiation at angles of 90° (sideways) and 180° (from the back), and it is very likely that the extreme values of response are at one of these 2 angles. Another limitation of this study is that only S–Cs source was used, which emits high energy photons. Additional testing should be performed in the future with low and medium energy X-rays, where deviations may be higher.

2.3.3. Energy dependence

The energy dependence testing was performed in the SSDL of ENEA, Italy using a 450 kV X-Ray source employing a high frequency H.V. generator (Bosello) and a 1 mm Be window of the X-Ray facility (X-Ray tube: COMET). The irradiation in terms of *H**(10) was done free in air, at a fixed distance in order to guarantee the desired dose rate and a homogenous irradiation of the dosemeters. The dimension of the field was 15 cm × 15 cm. At the same distance the homogeneity of the field was checked through measurements with a calibrated ionization chamber to guarantee an irradiation field with a photon fluence variability within 1%. Measured air-kerma values, at that fixed distance, have been converted to the required dosimetric quantity *H**(10) through ISO-4037 conversion coefficients. Five dosemeters from each system (total of 12 dosemeters types) were irradiated according to ISO 4037-3 at 8 different narrow series radiation qualities (N60, N80, N100, N120, N150, N200, N250 and N300) and also in S–Co, and S–Cs (ISO, 1999). The delivered dose was 2 mSv in all irradiations, with a measurement uncertainty of 9% (k = 2)

The energy dependence study was assessed as relative response of dosemeters at 10 radiation qualities within the mean energy range of 48 keV–1250 keV, normalized to the dosemeters indication at S–Cs (662 keV) radiation quality.

2.3.4. Natural environmental spectrum

The investigation of the response to a natural environmental spectrum was carried out in the Central Laboratory for Radiological Protection (CLOR) at two calibration stands equipped with different sources of gamma radiation S–Cs, S–Co and Ra-226. S–Cs and S–Co are accredited by the Polish Centre for Accreditation in accordance with the standard ISO/IEC 17025:2017. Irradiation with a sealed Ra-226 source (i.e. Ra-226 in equilibrium with its progeny) is used to assess the response of the detector to natural gamma radiation since Ra-226 and its daughters are radionuclides that naturally appear and its spectrum is similar to the wide gamma ray spectra occurring in the environment. Calibrations and irradiations are carried out in accordance with the ISO 4037-3 standard (ISO, 1999). The irradiations were made at a distance of 2 m from the source, with dose rates of 2.4 mSv/h for the S–Co source and 10.3 mSv/h for the S–Cs source (Fig. 2). The uncertainty for irradiations at the gamma calibration stand is 4% (k = 2).

The irradiations with Ra −226 were made at a distance of 1 m from the source with a dose rate of 0.28 mSv/h. The uncertainty for irradiations at the Ra-226 stand is 5% (k = 2). For each irradiation set-up, 5 dosemeters from each system were irradiated simultaneously free in air to a dose between 4.83 mSv and 5.04 mSv.

2.4. Calculation of uncertainties

The measurement uncertainties of the reference values were estimated by the respective calibration laboratories. Each laboratory estimated the measurement uncertainties of the reference values according to its own procedures. The largest contribution to the combined measurement uncertainty is the uncertainty of the conversion coefficient from air kerma free-in-air to ambient dose equivalent (*H**(10)). The recommended standard uncertainty of the conversion coefficient is 2% if the laboratory operates matched fields according to ISO 4037 standard (ISO, 1999). Other significant contributions are the uncertainty of the reference ionization chamber calibration factor, long term stability of the reference chamber and field inhomogeneity. Considering the large uncertainty of the conversion coefficient, uncertainties of temperature and pressure measurement, detector positioning, time measurements and other contributions can be neglected in most cases.

In this work the statistical uncertainty is estimated for each irradiation. At least 4 dosemeters were irradiated under each set of conditions (dose, angle, radiation quality). The statistical uncertainty was estimated by calculating the standard deviation of the readings of all irradiated passive dosemeters and multiplying it by an appropriate coverage factor to obtain a 95% confidence interval. Considering the small sample, coverage factors were calculated according to Student's T-distribution. An expanded statistical uncertainty below 10% is considered to be of sufficient quality for the purpose of this research.

3. Results and discussion

3.1. Dose dependent response

In general, the dose dependent response of passive dosimetry systems should be linear over a wide range of doses. At higher doses, saturation effects can lead to a supralinear or sublinear response when the dose is increased. In Fig. 3, the dose dependent response of different passive dosimetry systems using S–Co sources is shown. Results are presented as absolute response obtained by dividing the ambient dose equivalent mean value of 5 irradiated dosemeters by the reference value. The dose dependent response is measured in the dose range from 0.18 mSv to 1.2 Sv. The results show variations among different systems. For most of the systems, R^2 values between reference dose and readout value are close to 1 (0.9977–1.000). For one system (I) R^2 value is 0.9568. For most of the systems, (A, B, C, D, G, E, J) the calculated statistical uncertainty is below 10%. For System L, the statistical uncertainty is higher than 10% for all measured doses (up to 24%), except for one: for system I at four doses, the statistical uncertainty is between 13% and 22% and for systems F, K, H for two doses it is higher than 10%.

For the systems A, B, C, D, E, G, H, the difference between the measured values and the reference value is between 1% and 25% in the

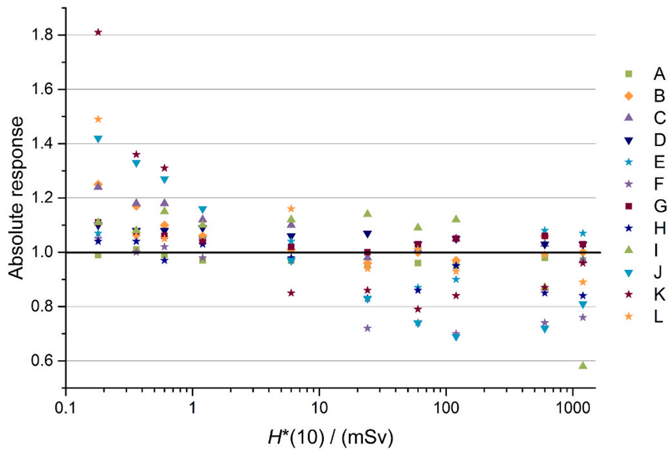

Fig. 3. Dose dependent response of different passive dosimetry systems in the dose range from 0.18 mSv to 1.2 Sv. The absolute response is expressed as the ratio of the mean measured dose value and the reference dose value.

whole measuring range. For some systems at certain doses the difference is higher – 30% for system F and even 80% for system K but with a statistical uncertainty higher than 10%. For system J, which is a film-based system, the difference in the response is higher, up to 42% in the whole measuring range. Most systems show only small dose dependence for doses below 0.1 Sv, which is quite sufficient for the purpose of long-term monitoring (even for highly contaminated areas).

3.2. Angular dependence

Results of the investigation of the angular dependence of passive dosimetry systems, using a S–Cs source, are presented in Fig. 4. Results for two TLD systems (K and L) are omitted from the graph because the statistical uncertainty is much higher than 10% (in some cases over 50%). Results are presented as the relative response for different angles of incidence normalized to the response for 0°. Relative responses for angles of 30°, 60° and 180° are within ±5% in 28 out of 30 cases, and the deviations from unity are significant only in 8 cases. However, all the tested systems show under-response for the angle of 90°, between −4% and −11%. In realistic conditions, during environmental radiation monitoring, it is very unlikely that a significant percentage of incident photons impacts the dosemeters at angles close to 90°, considering that the sources of photons are on (or partly in) the ground, due to potential contamination or in air. If the orientation of the dosemeter is horizontal, the contribution from secondary cosmic radiation is highest at small angles. Even if the dosemeter is positioned in vertical orientation it is very likely that the angular response is more or less averaged. Considering that the average angular response for the investigated systems is between −0.8% and −4.5%, this influence quantity is not very significant for the investigated passive dosimetry systems used for environmental dosimetry. It should be stated that the angular dependence was not investigated for low energy photons in this study, and that the results may be significantly different for some systems.

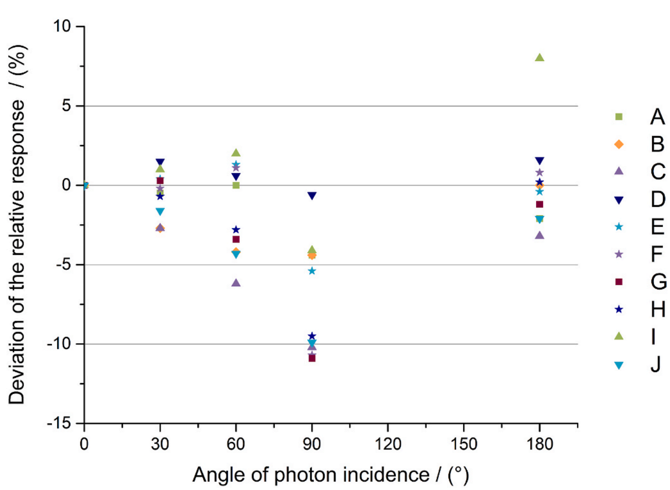

Fig. 4. Angular dependence of the response for the 10 different systems normalized to the angle of incidence of the photon radiation (S–Cs) of 0°. The deviation (in %) of the response ("relative response") is plotted for 5 different angles of incidence.

3.3. Energy dependence

The study of the energy dependence of the dosemeters' response was performed with 10 radiation qualities within an energy range of 48 keV–1250 keV (see section 2.3.3). This range covers the mandatory measuring range according to IEC 62387:2020 standard, which is 80 keV to 1.25 MeV. Photons with low energy (<80 keV) do not contribute significantly to the dose rate in the natural environment because they are shielded to a large extent by air and objects present in the environment. Therefore, with the exception of a few radionuclides, e.g. Am-241 (60 keV), the low energy region of the gamma spectra is not relevant for the total dose. Thus, a lower cut-off of the energy range at 80 keV, as defined by IEC 62387:2020, is normally sufficient for the environmental radiation monitoring. Due to this standard, the relative response of the dosemeters due to the radiation energy of the photons should be within the range limits from 0.71 to 1.67 (IEC 62387:2020) with respect to the standard calibration because otherwise the measured dose strongly depends on the spectrum of the photon radiation field. The results (Fig. 5a) of this study show that the energy dependence of the response, normalized to S–Cs, for three tested systems (A, G, H) is within these limits in the whole tested energy range (with the statistical uncertainty within 1%–10%.) (Fig. 5a) The response for four systems (B, C, I, K) performs slightly outside the limits (Fig. 5b). However, if the uncertainty of the reference values is being taken into account, these results are still satisfactory. System B is within the limits except for one energy, but with higher statistical uncertainty at some energies of up to 21%. For system K, the response is within the limits except at 100 keV, but with higher statistical uncertainties ranging between 17% and 26% for all tested energies. System D (Fig. 5c) shows a slightly under-response in the energy ranges between 48 and 118 keV, while for the system E (Fig. 5c) the results show a clear under-response and all the values are outside the limits. The energy response is normalized to S–Cs and the system E shows significant over-response to S–Cs (30%) which influences the other results. Considering that this system showed pronounced long-term instability in other tests, and that the irradiation in S–Cs has been made at a different time than the irradiation in X-ray qualities (due to technical problems), it is likely that the demonstrated energy dependence is due to the system's instability rather than the properties of the detectors themselves.

The results for systems F and J are not shown in the graphs due to their pronounced energy dependence in the entire investigated range of energies. System J, which is a passive dosimetry system based on film dosemeters, shows high overestimation (between 46% and 118%) of the response in the low-energy region (48 keV–83 keV) while in the higher energy region between 118 keV and 1250 keV an under-response of up to 80% is measured. System F shows a very high overestimation of the response in the whole energy range with a statistical uncertainty of 10%, except for two energies (65 keV and 118 keV) where the uncertainty was 13% and 24%, respectively. The relative response (normalized to S-Cs) varies between 2.0 for the highest energy to 9.05 for the lowest energy. The System F is a TLD based system whose dosemeters contain pairs of 2 detectors based on CaF_2: Mn and LiF: Mg, Cu, P material packed in a plastic bag. CaF_2:Mn is a material with high Z_{eff} (16.3), responsible for exhibiting a significant over-response, especially in the

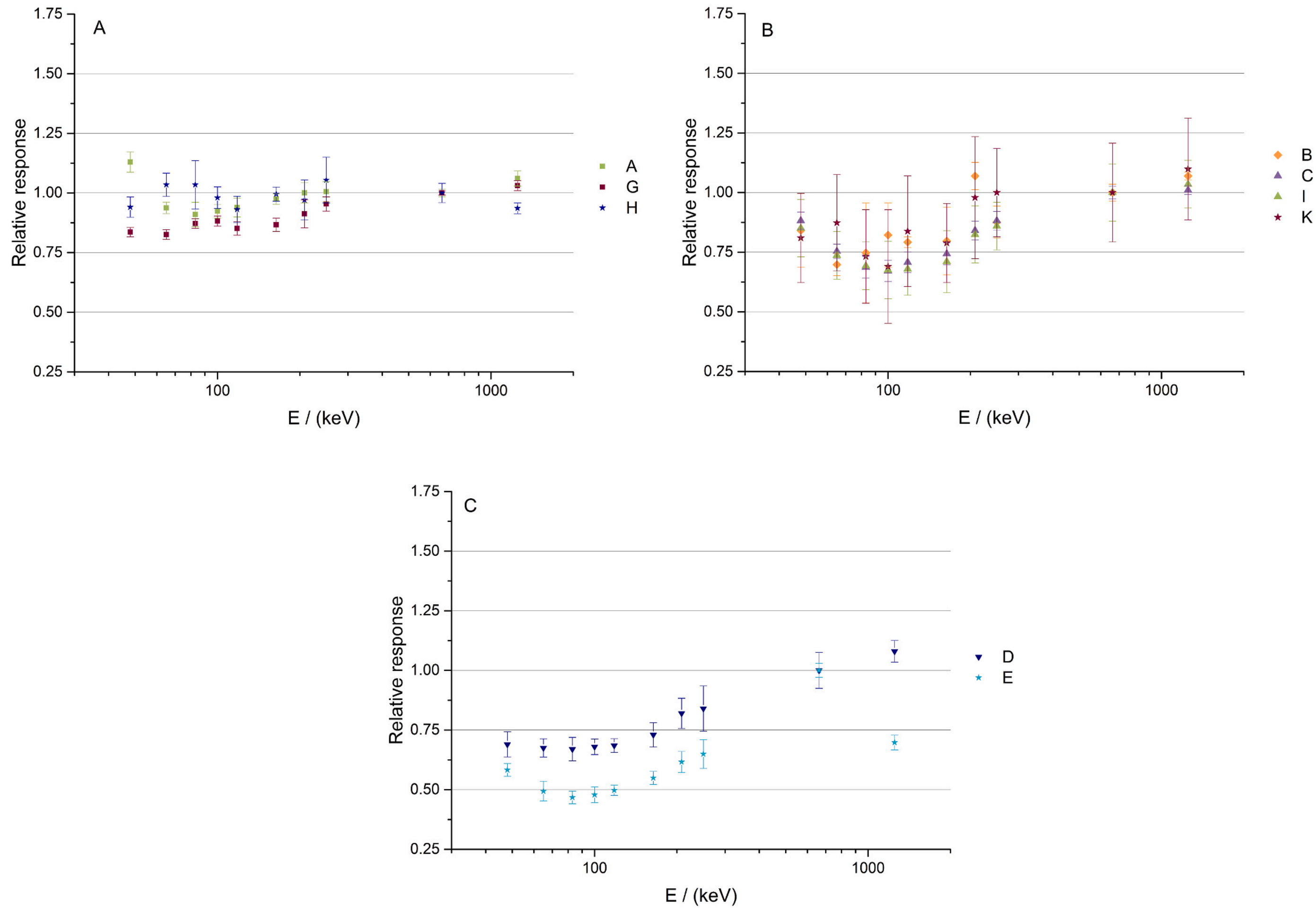

Fig. 5. (a–c) Relative energy response of different passive dosimetry systems normalized to S–Cs (i.e. 662 keV). The indicated uncertainties are statistical uncertainties.

low-energy region (McKeever et al., 1995). Also, the detectors are packed in holders without additional energy compensation filters. For environmental dosimetry this over-response is considered as a significant limitation. In principle, the over-response could be compensated by using energy flattening filters or/and corrected by dose algorithms. However, the use of algorithms can introduce an additional uncertainty into the dose estimation process which is not a trivial task in environmental dose calculations.

3.4. Natural environmental spectrum

Fig. 6 shows the results of the irradiation of passive dosimetry systems in a Ra-226 photon field. The results are normalized to the reference dose of 5 mSv. Ra-226 irradiation is used to assess the response of the detectors to natural gamma radiation. The majority of the systems show an under-response to Ra-226 but within 20%, except for the systems K and J which show an under-response of 33% and 53%, respectively and system F which show over-response by 21%. The statistical uncertainty U (mean) of the results is within 10% for the majority of the systems, except for the systems L, F, K and H (up to 14%).

3.5. Comparison of dosimetry systems for S–Co irradiations in different laboratories

In order to evaluate reproducibility, investigated passive dosimetry systems were irradiated by S–Co radiation quality in 3 different SSDLs. The radiation quality and angle of incidence were the same, and

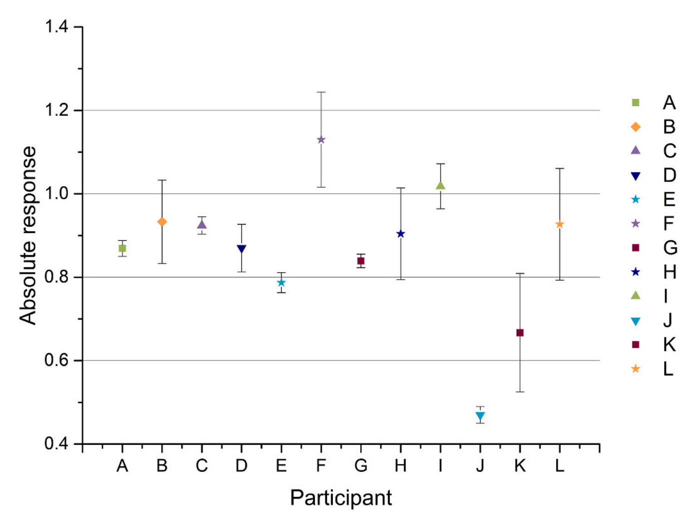

Fig. 6. Absolute response (expressed as mean measured value/reference value) of different passive dosimetry systems to the photon radiation of a sealed, radon tight Ra-226 source. The indicated uncertainties are statistical uncertainties.

delivered doses were similar. It should be noted that the irradiations were performed in intervals of several months and the individual passive dosemeters were not the same, even though the passive dosimetry systems were the same which could influence the results and variations. For most systems, the absolute response values to S–Co irradiations (Table 2) show very good consistencies and reproducible results for the irradiations in different standard laboratories (Lab1, Lab2, Lab3). The differences of the response values between irradiations of the same system are for the majority of the systems between 2% and 16% with a statistical uncertainty below 10%. However, in some cases, the absolute response varies significantly. For example, in case of system A and E, it varies between 1.29 to 0.94 and 0.77 to 1.05 respectively, which can't be explained by statistical uncertainty or by uncertainty of the irradiations in SSDLs. The passive dosimetry services were notified of the inconsistency, in order to check their procedures. The Systems K and L have a statistical uncertainty over 10%, similar to the other tests performed within this study.

Table 2
Comparison of dosimetry systems for irradiations in S–Co photon fields performed in different laboratories.

	Lab 1	Lab 2	Lab 3	Lab 3
applied dose	2 mSv	4.83 mSv	1.2 mSv	6 mSv
Participant	**Absolute response and (U_{mean}[a])**			
(A)	1.29 (3%)	0.94 (2.2%)	0.97 (2.3%)	0.97 (4%)
(B)	1.08 (2%)	1.01 (5.3%)	1.06 (4.2%)	1.01 (4.7%)
(C)	0.99 (2%)	0.96 (1.4%)	1.12 (3.2%)	1.10 (6.4%)
(D)	1.08 (5%)	1.04 (2%)	1.09 (1.9%)	1.06 (2.4%)
(E)	0.94 (3%)	0.77 (2%)	1.05 (1.6%)	1.04 (2.2%)
(F)	1.02 (4%)	1.18 (6.4%)	0.98 (1%)	0.97 (3.3%)
(G)	1.0 (2%)	1.00 (1.7%)	1.04 (1.1%)	1.02 (3.3%)
(H)	0.95 (2%)	0.97 (2.7%)	1.03 (3.5%)	0.98 (1.8%)
(I)	1.18 (4%)	1.13 (4.1%)	1.10 (7.1%)	1.12 (5.1%)
(J)	1.01 (5%)	0.84 (5.2%)	1.16 (2.8%)	0.97 (4.1%)
(K)	0.78 (21%)	0.80 (22.7%)	1.03 (10.1%)	0.85 (9.1%)
(L)	–	0.91 (9.7%)	1.06 (21.8%)	1.16 (24.5%)

[a] U_{mean} is the statistical uncertainty (the spread of the TLD readout values) of the absolute response (for a confidence level of 95%) and does not include the uncertainty of the reference value.

4. Conclusions

Passive dosimetry systems are suitable and usable for long-term radiation monitoring in the natural environment as well as after a nuclear and radiological event. In the aftermath of a nuclear or radiological accident, the additional dose rate (on top of the natural one) needs to be quantified. However, before their use for environmental radiation monitoring and also in case of an accident, investigations of the basic properties of the passive dosimetry systems are highly recommended. Within the Preparedness project, the dependence of the dosemeters' response as a function of dose, energy, angle of photon incidence and the response to a natural radiation field (approximated by the gamma radiation of a sealed Ra-226 source) were studied for 12 different passive dosimetry systems from 9 institutions. From all passive dosimetry systems tested within this study, OSL and RPL based systems show excellent performance in all tests.

For most of the tests, the calculated statistical uncertainties of the instruments' readings were below 10% but two systems based on TLD and one film system (J, L, K) show significantly higher statistical uncertainties.

The results show that the average angular response of the investigated systems (except for K and L) is between 0.8% and −4.5% and will not have influence on their use in environmental monitoring. This conclusion has the limitation that it was derived based on the irradiation in S-Cs field, which emits high energy photons, and the conclusion might not hold for some systems for the low energy photons.

All systems show acceptably low variations of the response to different dose values in a wide range of doses, (from 0.1 mSv to 100 mSv) taking into account the variations of the calibration factors among different radiation laboratories.

Four tested systems (A, G, H, I) show an energy dependence of the response within the limits defined by the international standard IEC 62387:2020 in the whole tested energy range while for several systems at some energies the response is a few percent outside these limits. However, these results can also be seen as satisfactory if the uncertainty of the reference value is taken into consideration. System E shows significant under-response in the whole tested energy range but the reason for this is a significant over-response to S–Cs.

The results for the two systems J, which is a film-based system, and F, which contains a detector with high Z_{eff} and a holder without an energy compensating filter, show a significant energy dependence in the whole investigated energy range. One of the systems (L) shows large statistical uncertainties and poor results in most of the tests. In addition, the response values of that system for irradiations in different laboratories (but under the same conditions) are not consistent. This is probably due to the inappropriate QA/QC procedures used by the laboratory, and/or individual dosemeter calibration factors. Therefore, before using this system in routine environmental radiation monitoring, the calibration of individual dosemeters should be checked and optimized.

Detector holders can also have an important influence on the energy and angular dependent response of passive dosimetry systems and currently there is no standard or recommendation about holders for the use in environmental radiation monitoring. Therefore, it is important to investigate the response for the combination of detector + holder. This is especially important as there is no standard or recommendation about holders and some institutions use their own home-made holders which may considerably deviate from the variety of commercially available holders. Following a nuclear event with released radioactive materials into the environment, very wide spectra, including low-energy photons (below 80 keV), can be expected. Therefore, it is important to use adequate filters over the detectors to compensate the energy dependence. Using several different detector types/materials can provide not only the dose but also information on the energy distribution of the photon radiation field and sometimes its mean photon energy.

As the use of different detectors, holders, calibrations, measurement procedures and uncertainties, leads to differences in the measured data, prior investigations and harmonization of passive dosimetry systems are necessary to achieve reliable and comparable dose measurements in Europe. In this study, these investigations have been performed for 12 different passive dosimetry systems and the results presented are a prerequisite for the use of these systems for a long-term environmental radiation monitoring in case of a nuclear or radiological emergency.

Funding

This project (16ENV04 Preparedness) has received funding from the EMPIR programme, co-financed by the Participating States and from European Union's Horizon 2020 research and innovation programme.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgment

The authors kindly acknowledge the great support of the following institutions by providing the respective dose values and associated uncertainties for the reported investigations: Central Laboratory of Radiological Protection (CLOR), Poland; ENEA-Radiation Protection Institute Italy; Institute for Occupational Health (IMI), Croatia; Jožef Stefan Institute (JSI), Slovenia; L.B. Servizi per le Aziende SRl (LBSA) Italy; Politecnico di Milano (POLIMI), Italy; Ruđer Bošković Institute (RBI), Croatia; Universitat Politècnica de Catalunya (UPC), Spain; Vinca Institute of Nuclear Sciences (VINS), Serbia. The authors also would like to express their gratitude to H. Dombrowski for his valuable discussions prior to and during the irradiation exercises to determine the basic properties of the passive dosimetry systems. The authors also like to acknowledge the excellent work of EURADOS WG3 in organizing intercomparison exercises for passive dosimetry systems on a more or less regular basis.

References

Budzanowski, M., et al., 2004. Long-term environment monitoring based on MTS-N (LiF:Mg, Ti) and MCP-N(LiF:Mg,Cu,P) thermoluminescent detectors. Radiat. Meas. 38 (4–6), 821–824.

Dombrowski, H., 2019. Preparedness Intercomparison of Passive *H**(10) Area Photon Dosemeters in 2017/2018. IC2017prep). JINST 14 P10008.

Dombrowski, H., Neumaier, S., 2012. Long-term PTB Intercomparison of Passive *H**(10) Dosemeters Used in Area Monitoring 2012 JINST 7 504017.

Duch, M., et al., 2017. Status of passive environmental dosimetry in Europe. Radiat. Meas. 106, 242–245.

Duch, M., Dombrowski, H., Hranitzky, C., 2021. EURADOS report 2021-02: overview of passive area dosimetry systems used in European countries. In: EURADOS Report 2021-02. EURADOS.

Iurlaro, G., et al., 2021. Study on the uncertainty of passive area dosimetry systems for environmental radiation monitoring in the framework of the EMPIR "Preparedness" project. Radiat. Meas. 142, 106543.

IEC, 2020 62387, 2020-01. Radiation Protection Instrumentation – Dosimetry Systems with Integrating Passive Detectors for Individual, Workplace and Environmental Monitoring of Photon and Beta Radiation. International Electrotechnical Commission, Geneva.

ISO, 2019. ISO 11929-1:2019, "Determination of the Characteristic Limits (Decision Threshold, Detection Limit and Limits of the Coverage Interval) for Measurements of Ionizing Radiation — Fundamentals and Application — Part 1: Elementary Applications".

ISO, 1999. International Standard ISO 4037-3, X and Gamma Reference Radiation for Calibrating Dosimeters and Dose Rate Meters and for Determining Their Response as a Function of Photon Energy – Part 3: Calibration of Area and Personal Dosimeters and the Measurement of Their Response as a Function of Energy and Angle of Incidence. Geneva.

McKeever, S.W.S., Moscovitch, M., Townsend, P.D., 1995. Thermoluminescence Dosimetry Materials: Properties and Uses. Nuclear Technology publishing, 1 870965191.

Nanto, H., et al., 2011. Environmental background radiation monitoring utilizing passive solid state dosimeters. In: Ekundayo, E. (Ed.), Environmental Monitoring. InTech, pp. 121–136. https://doi.org/10.5772/27382.

Neumaier, S., et al., 2019. Metrology for the mobile detection of ionising radiation following a nuclear or radiological incident . "Preparedness" Arh Hig Rada Toksikol 70, 62–68.

Preparedness Consortium, 2019. Publishable Summary for 16ENV04 Preparedness — Metrol-Ogy for Mobile Detection of Ionising Radiation Following a Nuclear or Radiological Incident. EURAMET website available on.

Ranogajec Komor, M., 2009. Passive solid state dosimeters in environmental monitoring. In: Aycik, G.A. (Ed.), New Techniques for the Detection of Nuclear and Radioactive Agents, NATO Security through Science Series B: Physics and Biophysics. https://doi.org/10.1007/978-1-4020-9600-6.

Ranogajec-Komor, M., 2003. Thermoluminescence dosimetry – application in environmental dosimetry. Radiation Safety Management 2, 2–16.

Saez-Vergara, J.C., 2000. Recent developments of passive and active detectors in the monitoring of external environmental radiation. Radiat. Protect. Dosim. 92 (1–3), 83–88.

Sarai, S., et al., 2004. Detection of self-dose from an OSL dosimeter and a DIS dosimeter for environmental radiation monitoring. J. Nucl. Sci. Technol. 41 (4).

Vekic, B., Ban, R., Miljanic, S., 2006. Secondary standard dosimetry laboratory at the Rudjer Boškovic Institute. Arh. Hig. Rada. Toksikol. 57, 189–194.