

TOTAL OCCUPATIONAL EXPOSURE DURING CHARACTERISATION, CONDITIONING AND SECURING OF RADIOACTIVE SEALED SOURCES: A NEW DOSIMETRIC CONCEPT USING ACTIVE ELECTRONIC DOSIMETERS

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Received in September 2008

Accepted in October 2008

Radiation dosimetry in protection against ionising radiation involves research of all possible pathways through which natural or man-made radioactive materials can contaminate a habitat and actually harm its biota. It also takes into account natural and artificial (man-made) electromagnetic ionizing radiation (γ and x radiation). This article presents a dosimetric study assessing exposure to man-made ionising radiation of local environment and total occupational exposure of two professional workers involved in characterisation, conditioning, and securing of unused radioactive sealed sources. The purpose of the study was to validate a new active electronic dosimeter (AED) of type ALARA OD and to develop a new monitoring method by tracing the external occupational exposure over real time. This method is used to continuously measure and record external radiation doses and, which is a novelty, establish dose rates receiving pattern as a function of real time. Occupational whole body dosimetric results obtained with AED were compared with results obtained with passive dosimetry (film badge and thermoluminescence). Air, dust, and silicon sand samples were analysed by gamma-spectrometry to estimate internal exposure of the two workers to ^{222}Rn due to inhalation or ingestion of dust and sand in indoor air. In order to establish total occupational exposure, control radon measurement was performed in the immediate environment and the external Hazard index (H_{ex}) was calculated.

KEY WORDS: AED, ALARA, effective dose, electronic dosimetry, hazard index, $H_p(10)$, low level waste, personal dosimetry, radiation protection, risk assessment

A modern ecological approach to protecting human life and environment from harmful effects of radiation requires a validated and integral concept of dosimetry. This concept does not distinguish humans from their environment and habitat (1, 2). It involves research of all possible pathways through which natural or man-made radioactive sources can contaminate habitats and harm biota. Dosimetric methods are followed by validated methods of risk assessment. Risk, as a measure of the probability of causing damage to life (3, 4), health, property, and/or environment, will

occur as a result of a given hazard (5), where hazard is regarded as a condition or physical situation with a potential for an undesirable consequences.

This article presents a dosimetric study assessing exposure to man-made ionising radiation of the local environment and total exposure to ionising radiation of two workers characterising, conditioning, and securing unused radioactive sealed sources. It also presents some features of a new active electronic dosimeter (AED), of type ALARA OD developed and utilised at the Institute for Medical Research and

Occupational Health (Zagreb, Croatia) to monitor and keep radiation doses in line with the *As Low As Reasonably Achievable occupational dose (ALARA)* policy (6, 7). For this purpose a new dosimetric method was developed for assessing the effective external occupational dose and environmental exposure burden (8, 9).

The study was done by a group of licensed professionals. The working and surrounding areas were constantly monitored with ionising radiation survey meters and were off-limits for anyone who was not under dosimetry surveillance. All licensed professionals carried film, thermoluminescence, and electronic dosimeters, including ALARA OD AEDs. Outdoor air was monitored using gamma-spectrometry. Additional sampling was done at the beginning and at the end of the study. Gamma-spectrometry was also used to analyse dust and sand particles from inside the storage containers. The results were used to assess possible internal radiation dose due to unexpected inhalation of contaminated dust originating from natural radionuclides, ^{222}Rn , or silicon sand bed in which a number of radioactive lightning rods and fire alarm units were stored. The lightning rods contained ^{60}Co or $^{152}\text{Eu}+^{154}\text{Eu}$, and the fire alarm units contained ^{241}Am .

The silicon sand bed, used to protect from radiation, filled a 2 m³ steel box placed at the far end of the storage area. As it contained only sealed sources, no contamination was expected from a direct contact unless sealed sources were extracted from their protective containers. A week after the study was over and the atmosphere in the storage area stabilised, a 24-hour radon indoor measurement was performed to determine radon concentration by calculating external Hazard index (H_{ex}) and to detect any non-conditioned radium source producing ^{222}Rn in the storage.

METHODS

We monitored continuous local background (LBG) ionising radiation (Figure 1) and performed control measurements of the local background ambient dose and dose rates outside the storage area. We also used Thermo Eberline FH 40 GL-10 survey meters with FHZ512A: 1.5" x 1.5" (NaI) and FHZ612 measuring probes. Measurements of ambient dose rate inside the storage area were not considered as measurements of natural background radiation (2), and are not described in this article. Inside the storage, external occupational

exposure to workers was measured, which took 10 consecutive days. AEDs can record doses that are almost up to three orders of magnitude lower than those detected by passive dosimeters. Their sensitivity is of the order of 100 counts per microsievert and can indicate doses as low as $\sim 15 \mu\text{Sv}$, which is within the natural background radiation range (7). AED ALARA OD is based on Geiger Müller (GM) tube. In order to accurately estimate occupational dose (in Gy or in Sv) (6) using AED ionising impulses to the GM tube of the AED were calibrated with radiation quality factor $Q=1$ for gamma rays. AED was utilised together with a film badge and thermoluminescence (TL) dosimeters to measure accumulated absorbed dose and to establish the dependence of the dose rate and real time of exposure to external gamma radiation. Film dosimeters used were AGFA Gevaert Structurix D2 GD10/GD20 films inserted into a PTW Freiburg film holder type 8621. TL dosimeters were Panasonic UD 802 AT with 4 elements, one pair $^6\text{Li}_2\text{B}_4\text{O}_7$ and one pair CaSO_4 .

AEDs ALARA OD continuously measured and recorded the doses and dose rates as a function of time, thus yielding a unique record of occupational doses for each worker and dose rates patterns at each specific working place. Time-dependent dose and dose rate patterns, total occupational dose, and duration of exposure were recorded for all workers involved in the study. ALARA OD has an alarm function which activates when dose rate exceeds a preset threshold value. This function is very important in procedures such as decommissioning of radioactive sources, where there is a possibility of acute exposure to high external doses due to container damage or other technical flaws.

The area near the storage had constantly been monitored over the past years and *in situ* gamma-spectrometric measurements were carried out. Samples of indoor dust during the study were collected and measured too. All samples were analysed in the laboratory using HP Ge and/or Ge(Li) detector (resolution 1.78 KeV on 1.33 MeV ^{60}Co , relative efficiency 16.8 %; resolution 1.56 KeV on 1.33 MeV ^{60}Co , relative efficiency 18.7 %). All samples were measured in 0.1-litre or 1-litre Marinelli beakers. Measurement time was 80,000 sec or higher. *In situ* gamma-spectrometric control measurements were carried out in front of the storage area door and at three sites in the surroundings, using a HP Ge Ortec detector (resolution 1.74 KeV on 1.33 MeV ^{60}Co , relative efficiency 21.6 %).

At these sites, measurements were performed before and after the characterisation of the stored sources. The measurements performed in front of the storage area were repeated after almost a year. Continuous measurements of external LBG exposure dose rate at the site located 100 m away from the storage area were additionally performed using ALARA OD AED 1 m above the ground, and were used as control measurements. Radon concentration measurements inside the storage area were performed using a Genitron Instruments Alpha GUARD radon monitor a week after the conditioning was completed, allowing for the ambient atmosphere in the storage area to stabilise. For this purpose a passive time-integrating method was employed.

RESULTS AND DISCUSSION

Exposure measurements in the environment

Figure 1 shows the measurements of natural LBG in 2006. The LBG measurement site is located at appropriate distance from the storage ventilation. Natural background ionising radiation at this site has been measured since 1959. The mean dose rate of 121.9 nSv h^{-1} was comparable to the survey meter dose rate measurements performed outdoors in front of the storage area, which means that no contamination reached the LBG measuring site during the study. Additionally, LBG was measured at the working site directly outside the storage area while the study was performed. Figure 2 shows these LBG values accumulated during the first 100 hours, starting from 7:00 PM one day before the study started. The results

are shown as cumulative dose because it is necessary to have reliable LBG data to be subtracted from the occupational cumulative dose $H_p(10)$ measured by AEDs worn by the workers. It is the same procedure of evaluating occupational dose as the procedure done for TL dosimeters, where LBG needs to be subtracted from TLD results. This was needed in order to compare the doses obtained by TLDs and AEDs.

The LBG values measured with AEDs are in agreement with the dose rates obtained with control survey meters (Figure 2).

Measurements of ambient dose rate inside the storage area were not considered as LBG (2). *In situ* gamma-spectrometric air measurements were carried out in front of the storage area almost a year before and a couple of months after the study. Figure 3 shows a small difference between the two measurements, which may be owed to differences in measurement performance, site location and atmospheric conditions rather than to any contamination. Figure 4 shows the measurements at the monitoring sites performed during the study.

The variation of dose rate produced by naturally occurring radionuclides is similar to measurements at the LBG site obtained over three years (Figure 5).

Figure 6 shows the results of gamma-spectrometry of mixed samples of dust and silicon sand from the protective bed inside the storage area. Man-made radionuclides are of special interest because they could be inhaled with dust, and contribute to the internal exposure component of the total occupational dose. The results in Figure 6 and Table 1 show that only traces of ^{60}Co , ^{241}Am , and ^{137}Cs (measurement of $^{152}\text{Eu}+^{154}\text{Eu}$ was in the range of detection level) were found in the sand bed samples, probably because

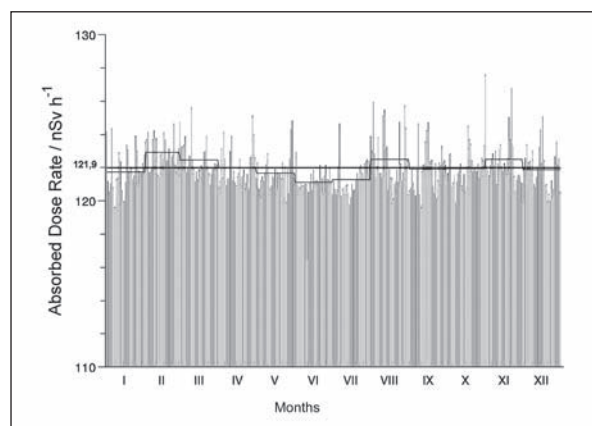


Figure 1 Variations of natural background radiation in 2006 at the local background radiation (LBG) monitoring station 100 m from the storage area.

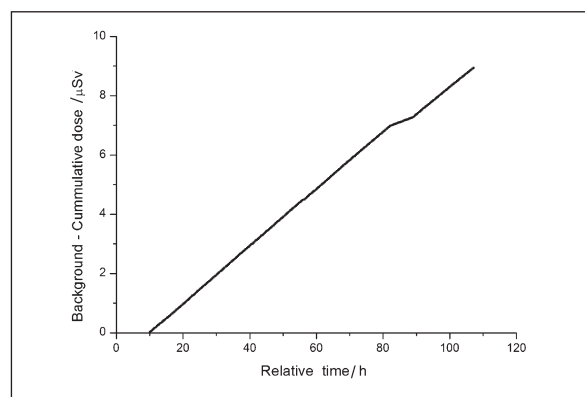


Figure 2 Local background radiation during the first five working days of the study, measured with AED at LBG monitoring station outside the storage in July of 2006.

some of the sealed sources had no original protective container, neither they were placed into plastic tubes but were put directly into the sand. Higher activity of all radionuclides in the upper sand layer confirmed slight contamination of the sand (Table 1) from the sources without protective container that were placed beside the plastic tubes - stored directly in plain protective sand bed. In contrast, the activity of natural radionuclides in sand bed samples was within the background range as expected (1, 2). The external Hazard index, so called H_{ex} , was lower than 1, suggesting that inhalation would not contribute to the total occupational dose (5). Radon was measured inside the storage area after the study was completed. We also wanted to find out if there was an uncharacterised radium source and if there was, to isolate it from all other sources. The results are shown in Table 2. The high radon value in air was measured only at the site inside the storage in which the characterized radium sources were finally placed. No other area was contaminated with radium, which means that the work was successful and that the indoor storage area was clean.

Table 1 Activity concentration of non-natural radionuclides in silicon sand samples taken from the upper and middle protective bed layer

Radionuclides	Activity concentration / Bq kg ⁻¹	
	Upper layer	Middle layer
¹³⁷ Cs	0.247±0.032	0.201±0.028
⁶⁰ Co	10.93±0.13	0.776±0.055
²⁴¹ Am	117.84±0.25	22.388±0.172
¹⁵² Eu	< (0.1269±0.0618)	< (0.1602±0.0780)
¹⁵⁴ Eu	< (0.1188±0.0583)	< (0.0774±0.0533)

Table 2 In situ gamma-spectrometric measurements in silicon sand samples taken from the upper and middle protective bed layer and external Hazard index (H_{ex})

Radionuclide	Activity concentration / Bq kg ⁻¹	
	Upper layer	Middle layer
²³⁸ U	17.61±4.35	20.00±4.37
²³⁵ U	0.983±0.081	0.781±0.053
²³² Th	13.84±0.50	14.14±0.42
²²⁶ Ra	14.897±0.913	14.48±0.91
²¹⁰ Pb	13.42±1.50	11.57±1.13
²²⁸ Ra	13.84±0.50	14.14±0.42
⁴⁰ K	254.23±1.29	253.8±1.29
H_{ex} / Relative number		
H_{ex}	0.14655	0.14649

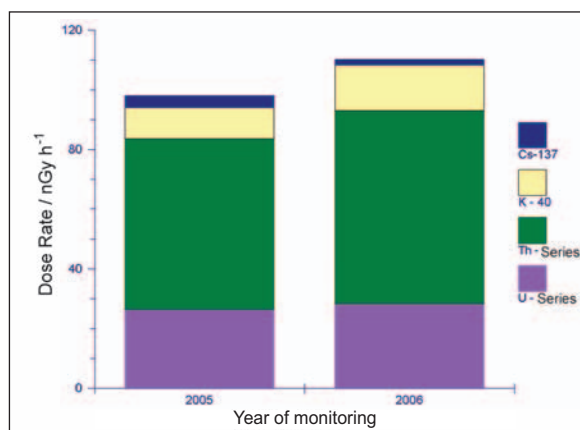


Figure 3 In situ ambient air gamma-spectrometry at location in front of the storage area before and after the study

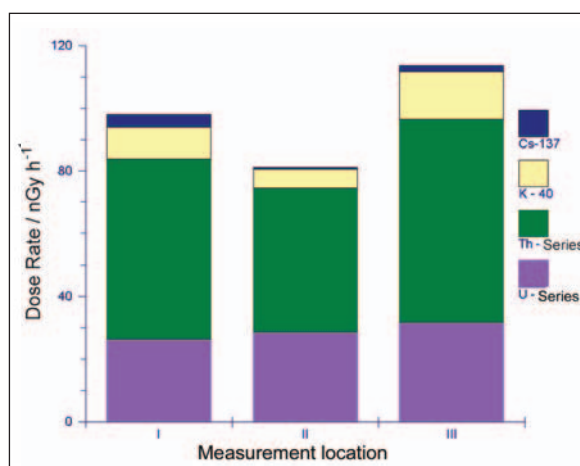


Figure 4 In situ ambient air gamma-spectrometry at four locations: I – outdoors, in front of the storage area; II – 60 m from the storage; III – 20 m from the storage; LBG monitoring station – 100 m from the storage

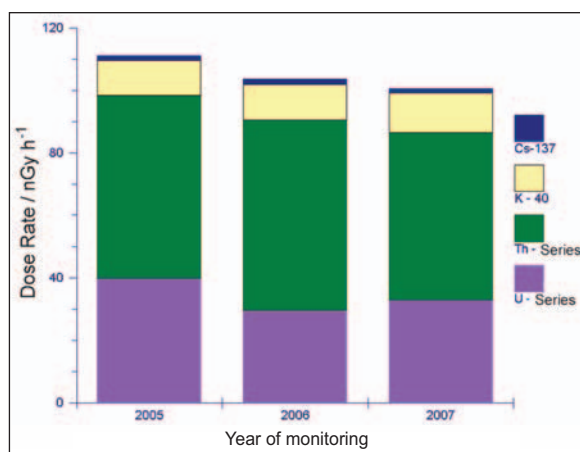


Figure 5 In situ ambient air gamma-spectrometry at the LBG radiation monitoring station obtained over years 2005-2007

The internal hazard index for radon in the storage area shows that its contribution to the total occupational dose (internal dose component) was

about 1 % (5). AEDs were measuring the external gamma component of radon radiation. The values are incorporated in a LBG from inside the storage which are shown in Figures 7-10. The dominant contributor to the total occupational dose was exposure to external γ radiation from sealed sources.

Personal occupational exposure

Here we also present the occupational burden of two workers characterising sealed sources and cleaning the temporary storage area and its immediate outdoor environment. Total exposure consisted of exposure to local natural background radiation, occupational external exposure to gamma radiation produced by old and/or unused sealed sources, and of possible internal exposure to ^{222}Rn or other isotopes inhaled with contaminated air, dust, and silicon particles from the protective sand bed.

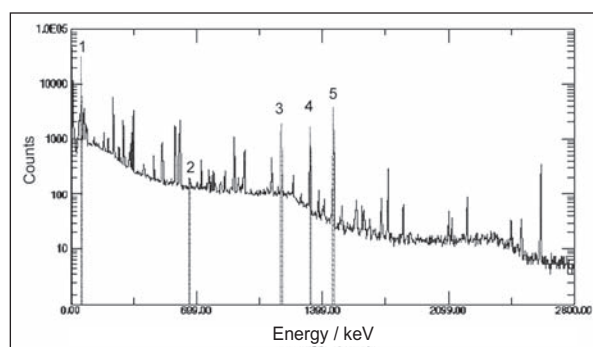


Figure 6 Gamma-spectrometry of collected mixed samples of dust and silicon sand from the protective bed inside the storage area. 1- ^{241}Am at 59.54 KeV; 2 - ^{137}Cs at 661.62 KeV, 3- ^{60}Co at 173.23 KeV, 4 - ^{60}Co at 1332.51 KeV; and 5 - ^{40}K at 1460.75 KeV.

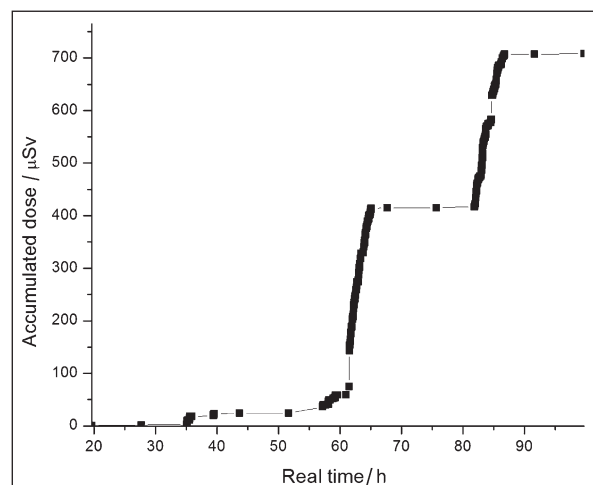


Figure 7 Four-day ALARA OD AED no. 220 readout. The accumulated equivalent dose for worker 1 was 708.8 μSv . AED was started at 19:39:41 PM and read after four days at 9:00:00 AM.

Figure 7 shows ALARA OD AED readout for a worker who worked in the area for four days. His accumulated equivalent dose was 708.8 μSv . The figure shows accumulated dose vs. real time in hours. AED was started at 19:39:41 PM and was read after four days at 9:00:00 AM. Higher accumulated doses were recorded only during work in the storage area; otherwise the AED recorded only LBG.

Figure 8 shows the dose rate vs. real time for the same worker. The dose rate peaks show the exact time of occurrence and duration of exposure. They form the exposure dose rate pattern showing exact real time when exposure occurred and the fragmented duration of the exposure. AED two-day readout of the second worker are shown in Figure 9. The accumulated equivalent dose for these two days equalled 308.2 μSv . The graph shows accumulated dose vs. real time in hours. AED was started at 9:42:54 AM and read after two days at 9:38:42 AM. Figure 10 shows dose rate peaks with the exact time of occurrence and duration of exposure. This worker was in contact with a sealed source radiating higher doses a few minutes before 3:00 PM on the first day. On the second day he worked outside the storage area, which is confirmed by AED reading only LBG on that day (Figures 9 and 10). This novelty feature, which allows fragmented occupational personal dose readouts, is not achievable with passive dosimeters (Figure 10). It makes it possible to work out the exact real time when the exposure occurred. This exact real time data will form exposure dose rate time pattern (Figures 8 and 10) which will be different for each of the two workers and show which one was more often near or in contact with sources.

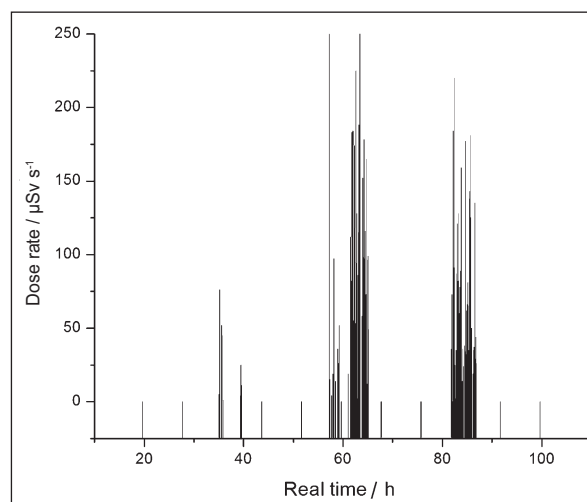


Figure 8 Four-day ALARA OD AED no. 220 readout. Dose rate for worker 1 vs. real time in hours.

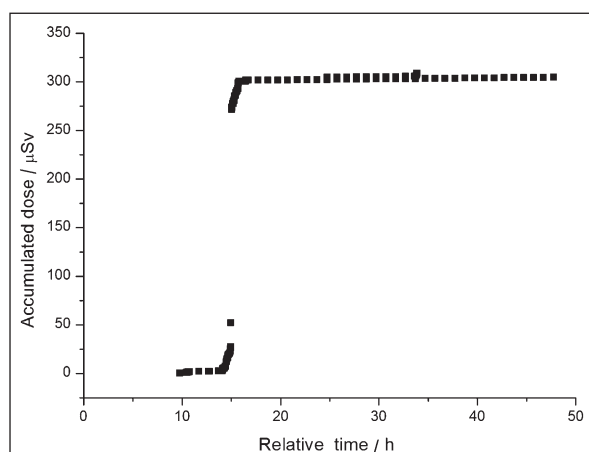


Figure 9 Two-day ALARA OD AED no. 204 readout. The accumulated equivalent dose for worker 2 was 308.2 μSv . AED was started at 9:42:54 AM and read after two days at 9:38:42 AM.

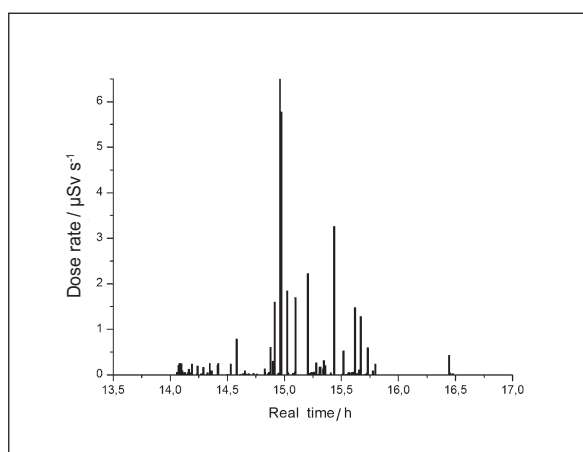


Figure 10 Dose rate for worker 2 vs. real time. The peaks show the exact real time of occurrence and duration of occupational exposure.

Table 3 shows radon concentrations at three locations. Higher radon concentrations were found only at one location inside the storage, that is, at the place of the conditioned radium source container. This means that no radium source outside the container was left unconditioned.

Additionally, Table 4 shows the results of regular and additional passive personal dosimetry expressed as total occupational dose ranges and dose rate ranges over all 10 working days.

CONCLUSION

ALARA OD AED can continuously measure LBG and at the same time record any radiation higher than LBG (usually 2 to 3 times the LBG). ALARA OD AED will automatically record all segments of occupational external gamma radiation. It can produce a detailed record of any single contact with a radiation source and show a pattern of the frequency and duration of handling radiation sources outside

Table 3 Radon concentrations inside the storage area.

	Location		
	Site near final container placement inside storage	Cleaned site inside storage	Reference indoor value measured 100 m from the storage
Radon concentration / Bq m^{-3}	11400 to 14000	400 to 620	360 to 440

Table 4 Total occupational equivalent dose ranges and dose rate ranges received by two workers over the 10 working days of the study

		WORKER	
		No. 1	No. 2
Total dose range Hp(10) / mSv	Film (regular dosimetry) one-month readout	0 to 14.30	0 to 0.40
	TLD (regular dosimetry) one-month readout	0 to 20	0 to 0.37
	TLD (additional dosimetry) 10-day readout	0.70 to 20	0 to 0.32
	ALARA OD AED 10-day readout	0.03 to 20	0.03 to 1.2
Dose rate range / $\mu\text{Sv s}^{-1}$	ALARA OD AED 10-day readout	LBG to 320	LBG to 20

LBG – local background

the protective container, possible number of sources manipulated, or any other type of contact and contact duration with sources. The dose rate data presented here describe only four (first worker) and two days (second worker) of work, but they clearly show how much time each worker spent in the vicinity of a radiation source. This is a novel feature in describing occupational exposure, and it could find application in special (nuclear, radwaste), additional (nuclear medicine), or even regular personal dosimetry in the future (7-10). Together with the hazard index (H_{ex}) obtained for possible ^{222}Rn inhalation of air or dust particles, it is possible to estimate the total radiation exposure burden for occupationally exposed workers performing their duties in dosimetrically complex environments.

Acknowledgments

This work was supported by the Ministry of Science, Education and Sports of the Republic of Croatia as a part of the research project "Environmental Radioactivity and Radiation Protection" (grant no. 022-022882-2335).

The authors thank to all participants involved in the study. Our special thanks goes to our technician Jasminka Senčar and her invaluable help in performing gamma-spectrometry.

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Sažetak

UKUPNA PROFESIONALNA IZLOŽENOST TIJEKOM KARAKTERIZACIJE, KONDICIONIRANJA I OSIGURAVANJA ZATVORENIH RADIOAKTIVNIH IZVORA: NOVI DOZIMETRIJSKI KONCEPT UPORABOM AKTIVNIH ELEKTRONSKIH DOZIMETARA

Dozimetrijski koncept zaštite od ionizirajućeg zračenja podrazumijeva istraživanje svih mogućih putova kojima radioaktivni materijal nastao u prirodi ili kao posljedica ljudskog djelovanja može ući u okoliš i utjecati na biotu ili onečistiti habitat oštećujući biotu. Taj koncept također uzima u obzir prirodno i umjetno (stvorenno ljudskim djelovanjem) elektromagnetsko ionizirajuće zračenje (γ i X-zračenje). U ovom radu predstavljena je dozimetrijska istraživačka studija kojom se istražuje i procjenjuje ukupna profesionalna izloženost dvaju radnika te izloženost lokalnog habitata ionizirajućem zračenju. Studija je obuhvatila karakterizaciju, kondicioniranje i osiguravanje nekorištenih, uskladištenih zatvorenih radioaktivnih izvora u cilju potvrđivanja i validacije novog aktivnog elektronskog dozimetra (AED) tipa ALARA OD te razvoj nove metodologije praćenja vanjske profesionalne izloženosti u vremenu. Kontinuirano mjerenje i snimanje primljenih vanjskih doza te kao novitet, brzine doze kao funkcije vremena, daju novu i jedinstvenu snimku profesionalnih doza i uzoraka primanja brzine doze na danome radnome mjestu. Rezultati mjerenja profesionalne doze za cijelo tijelo dobiveni uporabom AED-ova uspoređeni su s dozimetrijskim rezultatima [^{10}Hp] dobivenim uporabom pasivnih dozimetara (filmskih i termoluminiscentnih dozimetara). Provedene su gamaspektrometrijske analize zraka, prašine i silikatnog pijeska te su dobiveni podaci iskorišteni za procjenu moguće unutarnje izloženosti radnika ^{220}Rn inhalacijom zraka ili ingestijom prašine i pijeska. Radi određivanja ukupne profesionalne izloženosti bilo kojeg radnika uključenog u ovaj projekt, provedena su kontrolna mjerenja radona i izračunan je Hazard index (H_{ex}).

KLJUČNE RIJEČI: AED, ALARA, efektivna doza, elektronska dozimetrija, hazard indeks, ^{10}Hp , niskoradioaktivni otpad, osobna dozimetrija, procjena rizika, profesionalna izloženost, zaštita od zračenja

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