RELATION OF THE AMBIENT DOSE EQUIVALENT RATE IN AIR AND THE AMOUNT OF PRECIPITATION DURING ENVIRONMENTAL MONITORING IN THE VICINITY OF NUCLEAR FACILITIES

by

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Environmental radiation monitoring involves measuring the level of radioactive contamination of the air, including the relevant meteorological measurements at the micro-location type of measurements, which are essential for evaluating the extent of environmental factors and effectively managing the exposure of the population and the impact on the environment. These measurements are crucial in ensuring a comprehensive understanding of the ecological conditions and facilitating informed decision-making to safeguard the well-being of communities and ecosystems. This paper shows the relation between the change of ambient dose equivalent $H^*(10)$ rate in the air and precipitation due to washing out and rainout in the atmosphere. Measurements were made in the vicinity of nuclear facilities within the public company Nuclear Facilities of Serbia at the weather station mounting pole 114 m above sea level. To illustrate the relationship between the measured ambient dose equivalent $H^*(10)$ rate and precipitation levels, a variation of these values was employed specifically during the most rain-laden months in 2019 and 2020. Besides, an interlaboratory comparison was conducted to ascertain the system's operational validation. A thorough examination of this ratio distinctly reveals the impact of heavy rainfall on the ambient dose equivalent $H^*(10)$ rate, thereby rationalizing the observed elevated values. Importantly, these heightened readings were not attributed to any inadvertent release of radioactive effluents from nuclear facility operations in this particular instance.

Key words: monitoring, environment, meteorological parameter, precipitation, nuclear facility

INTRODUCTION

Protection of the environment from ionizing radiation is a set of measures that prevent the harmful effects of ionizing radiation in the environment and eliminate the consequences of that radiation, as well as keeping evidence of materials and raw materials that increase the concentration of radionuclides above the prescribed limits in the technical-technological process [1].

The public company Nuclear Facilities of Serbia (PC NFS) is the only operator of nuclear facilities and the holder of a license to carry out nuclear activities in the country. The PC NFS operates the research nuclear reactor RA, which is in preparation for decommissioning activities, the experimental zero-power nuclear reactor RB, two old radioactive waste storage facilities with the nuclear license for the final shut-down, two new radioactive waste storage facilities in operation (Hangar H3 and secure storage SS), a waste processing facility in preparation for trial-run, and a closed uranium mine Gabrovnica in eastern Serbia.

Following current legislation, PC NFS monitors radioactivity in the environment in the vicinity of nuclear facilities and examines the level of external radiation by the ambient dose equivalent $H^*(10)$ rate measurement in the air, the relevant meteorological measurements, and modeling the dispersion of radioactive pollutants in the boundary layer of the atmosphere.

Monitoring of radioactivity in the environment is carried out to assess the population's exposure to ionizing radiation, both from natural and artificial sources. Natural radioactivity as an integral part of our environment continues to be the main source of radiation exposure and impact on the environment, with 85.5 % of the average radiation dose received by the population. In the boundary layer of the atmosphere, natural radioactivity comes mostly from radon ^{222}Rn

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and thoron ^{220}Rn, which are present in the atmosphere as part of the natural decay chains of uranium and thorium. The progeny of these radionuclides spread in environmental air and are removed from the atmosphere by processes such as dry and wet deposition. With dry deposition, progeny falls to the ground due to gravity or airflow. This paper deals with the wet deposition process occurring due to the washing of progeny by precipitation.

The climate of the Vinča area belongs to the moderately continental type with four seasons. The autumn period is longer and warmer than the spring period. Summers are warm, while in winter there is snow cover. Spring is short and rainy, and summer comes quickly. The warmest months are July and August, while the coldest is January. Data covering the period from 1958 to the present in terms of monitoring meteorological parameters at the Vinča micro-location are available.

The annual average amount of precipitation was 684 mm from 1961 to 1990. The highest amount of precipitation occurs in June, with an average monthly amount of precipitation of 90.4 mm. The driest month is October, with a mean monthly rainfall of 40.3 mm. The location and coordinates of the meteorological station are shown in tab. 1. and fig. 1.

During the rainy period, radioactive elements are washed out from the atmosphere to the ground, and a temporary increase in the ambient dose equivalent $H^*(10)$ rate is observed. Therefore, it is important to distinguish in real time whether possible increases and changes in the measured values of ambient dose equivalent $H^*(10)$ rate in the air result from changes in meteorological conditions or the occurrence of a radiation or nuclear accident.

Apart from precipitation, the ambient dose equivalent $H^*(10)$ rate in the air generally depends on geographical location, mainly influenced by altitude (cosmic radiation), the radioactive content of local rock formation and soil (naturally occurring minerals with significant concentrations of uranium, thorium, and potassium), as well as by nuclear accidents and tests in the past (contamination by artificial radionuclides).

Table 1. Co-ordinates of the measuring station

Co-rdinates of the measuring station
Longitude: λ = 20°35'59.85"E
Latitude: φ = 44°45'38.55"N
The altitude of the object: 114 m

The accident at the Chernobyl nuclear power plant (1986) raised awareness of possible nuclear accidents as well as long-distance transportation of nuclear material depending on meteorological conditions. This led to the development of a system for continuous measurement of the dose rate of gamma radiation in the air, as well as early warning of a radiation accident in most countries.

Nuclear facilities in the PC NFS are an artificial source of radiation. Continuous measurement of ambient dose equivalent $H^*(10)$ rate in the air as part of the monitoring of radioactivity in the environment around nuclear facilities is carried out to fulfill the safety and security conditions for the operation of nuclear facilities under the Law [1] and Rulebook [2]. The tests are carried out by an accredited method following the requirements for the competence of testing laboratories according to the SRPR ISO/IEC 17025:2017 standard (Testing the level of radioactive air contamination by continuous measurement of the strength of the ambient dose equivalent: EPA HASL 300:1997), by the Conformity Assessment Body of Serbia [3, 4].

Routine interlaboratory analyses are carried out to enhance the analyses of precipitation's influence on

Figure 1. Location of the measuring station

radiation conditions in both work and living environments and to evaluate their relation.

An interlaboratory comparison of continuous measurement of the ambient gamma dose rate equivalent was performed.

MEASUREMENT AND METHODS

As part of the monitoring of radioactivity in the environment, continuous measurement of the ambient dose equivalent rate of gamma radiation in the air is carried out at five locations in the vicinity of nuclear facilities and one at a remote location, Vodovod, in the village of Vinča, which is part of the system of early warning of emergencies at the state level, which is within the competence of the Directorate for Radiation and Nuclear Safety and Security of Serbia. The data continuously collected at the latter location, are also part of the European Radiological Data Exchange Platform (EURDEP) [5]. The coordinates and location of the measuring station are given in tab. 1 and fig. 1.

The EURDEP platform also shows data on the environmental ambient gamma dose rate equivalent in the countries surrounding the Republic of Serbia. This is crucial as monitoring of the dose rate in the neighboring countries where nuclear power plants are installed enables the tracking of potential increases in the ambient dose equivalent $H^*(10)$ rate. Depending on the prevailing weather conditions and atmospheric currents, the airborne contaminant cloud may extend to our country as well.

Data on the ambient dose equivalent $H^*(10)$ rate measurements in the air are collected continuously for 24 hours, and the half-hourly values are displayed on the main monitor in the control room of the Department for Radioactivity Examination and Environmental Protection of the PC NFS. In addition to light and sound signaling, the system has been improved with the help of the eEMIS application and SMS notification in case of an increased ambient dose equivalent $H^*(10)$ rate above the defined threshold, which in this case is approximately double the value of the natural background (200 nSvh^{-1}) [6]. Also, this value is approximately double the value of the measured background during system commissioning, represents the new reference initial state, and is a tradeoff between sufficient system sensitivity and low false alarm events. Company AMES from Slovenia developed the eEMIS application.

The SMS notification about the increased dose rate reaches the phone numbers of the persons authorized for radiation measurement, as well as the person in charge of radiation safety in the PC NFS.

Previous practice has shown that notification of increased values of the dose rate was received only due to increased values during rainfall, which was confirmed and proven in regular reports [7].

Continuous ambient dose equivalent $H^*(10)$ rate measurements were obtained using MFM203 monitors. MFM203 is a portable instrument for monitoring gamma radiation in the environment. It autonomously performs all the necessary functions of a self-sustained local monitor. When connected to a data communication link, it operates as a smart field unit of the real-time automatic early warning network. The two energy-compensated Geiger-Müller-based probes of different sensitivities and measurement ranges, MFM203 can cover dose-rates from the normal background to accidental levels.

Two probes are positioned at 1.3 m above the grass surface. The site is elevated 114 m above sea level, which allows for unhindered airflow around the probes.

The measuring range of probes is from the background level of ambient gamma dose rate equivalent (50 nSvh^{-1}) up to accidental levels (1 Svh^{-1}). The energy response of the probes is from 50 keV to 2 MeV, linear in the energy interval from 60 keV to 1.3 MeV within ±20 %. The Dose rate display range is in Sievert per hour units, limited to a five-digit decimal significand (mantissa) and a one-digit decade exponent (XXXXXe+Y) [8].

The gamma dose rate monitor with probes (A and B) is presented in fig 2.

From the radiation protection point of view, it is important to determine the relation between ambient dose equivalent $H^*(10)$ rate in the air and meteorological parameters. Meteorological parameters, *i.e.*, air temperature at different reference heights, air pressure, the wind at three levels, precipitation, relative humidity of the air, and solar radiation, were obtained by automated measuring devices as part of the automated meteorological station set up on the meteorological weather station mounting pole of the 40 m of height, on 114 m above sea level [9].

Figure 2. Multifunctional gamma dose rate monitor MFM203 with probes

The structure of the meteorological tower was renewed in 2008, and sensors were placed on three levels at the meteorological tower: 2 m, 10 m, and 40 m. An automated weather station with sensors (AMS156), made by the Slovenian company AMES, is presented in fig. 3. and fig. 4.

An automatic electric rain gauge with a heater fig. 5, whose resolution is 0.2 mm [9], placed 1 m above the ground, obtained data on the amount of precipitation.

All collected data and measured values are represented and analyzed by the application eEMIS.

The continuous measurement results of the ambient dose equivalent $H^*(10)$ rate in the air are shown in units of $nSvh^{-1}$. The combined measurement uncertainty of the results is estimated to be 28.64 % with a confidence level of 95 % ($k = 2$), for the common effluent energy range (60 keV to 1.3 MeV), by the internal documents of PC NFS [10].

RESULTS AND DISCUSSIONS

Graphical presentations of ambient dose equivalent $H^*(10)$ rate during significant rainy periods in

Figure 3. Automated meteorological weather station

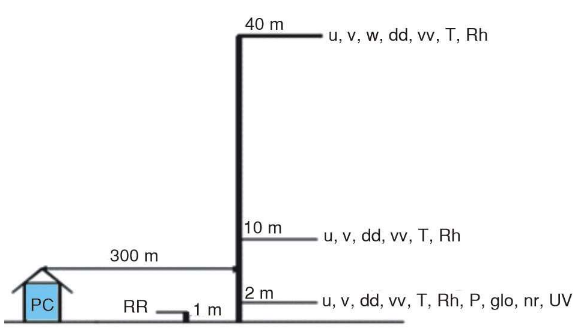

Figure 4. Scheme of the meteorological sensors at the meteorological tower

Figure 5. Rain gauge with heater

2019 and 2020, when the relation with precipitation is particularly evident, are shown in figs. 6 and 7 for December 2019, and in figs. 8 and 9 for February 2020, respectively.

The Pearson correlation coefficient was computed to determine the atmospheric natural radioactivity dependence on precipitation.

The Pearson correlation coefficient quantifies the extent of dependence between two physical measures, providing a value within the inclusive range of +1 to –1. It measures the strength and direction of a linear relationship between two variables [7].

The following criteria were used for Pearson's correlation coefficient: uncorrelated (for values that are between –0.09 and 0.09), low correlated (for values that are between –0.3 and –0.1or between 0.1 and 0.3), medium correlated (for values that are between –0.5 and –0.3, or between 0.3 and 0.5), and strong correlated (for values that are between –1.0 and –0.5 or between 0.5 and 1.0) [7].

The Pearson correlation coefficients were computed using Microsoft Office Excel by the function Pearson. Values of 0.82 for December 2019 and 0.43 for February 2020 were obtained after calculation.

Observing the graphical presentations and computed Pearson's coefficient, a medium to strong correlation between precipitation and ambient dose equivalent $H^*(10)$ rate becomes evident. It is apparent that the rise in ambient dose equivalent $H^*(10)$ rate was not a result of a specific radiological accident but occurred notably during the rainiest months in 2019 and 2020.

It is important to know that the shown increases do not exceed the value of 200 $nSvh^{-1}$, which is the value of the set alarm threshold.

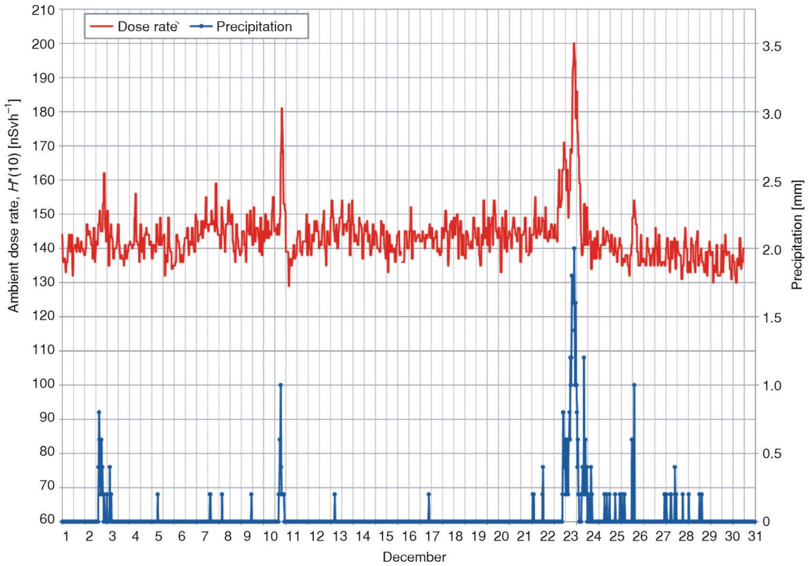

Figure 6. Ambient dose equivalent $H^*(10)$ rate and precipitation during December 2019

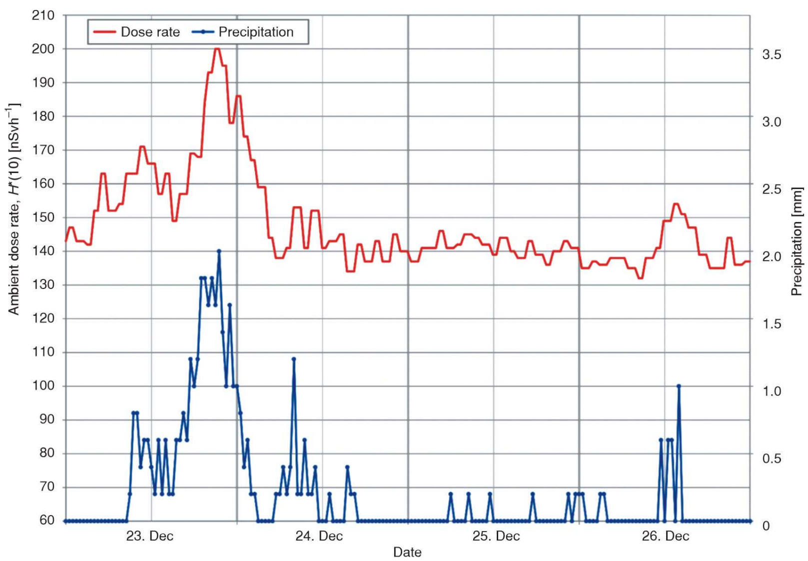

Figure 7. Ambient dose equivalent $H^*(10)$ rate and precipitation during December 2019 with special analysis of the rainiest period (23.12.2019-26.12.2019)

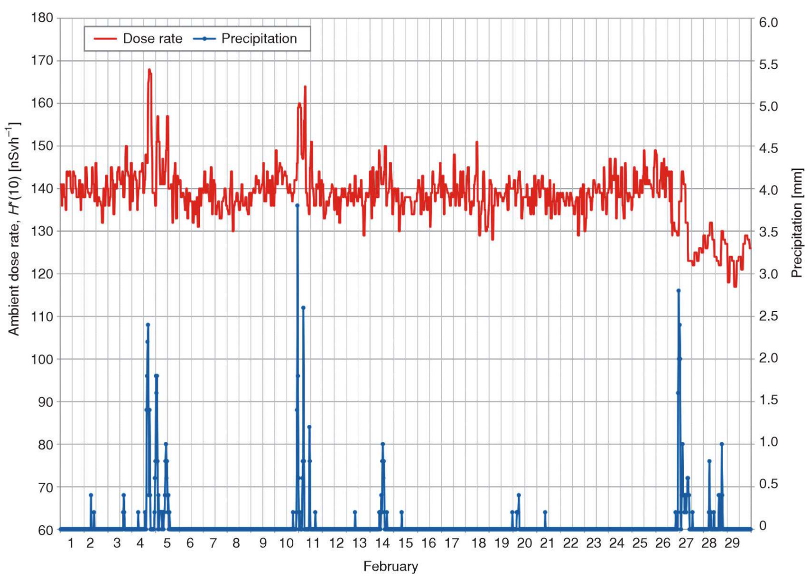

Figure 8. Ambient dose equivalent $H^*(10)$ rate and precipitation during February 2020

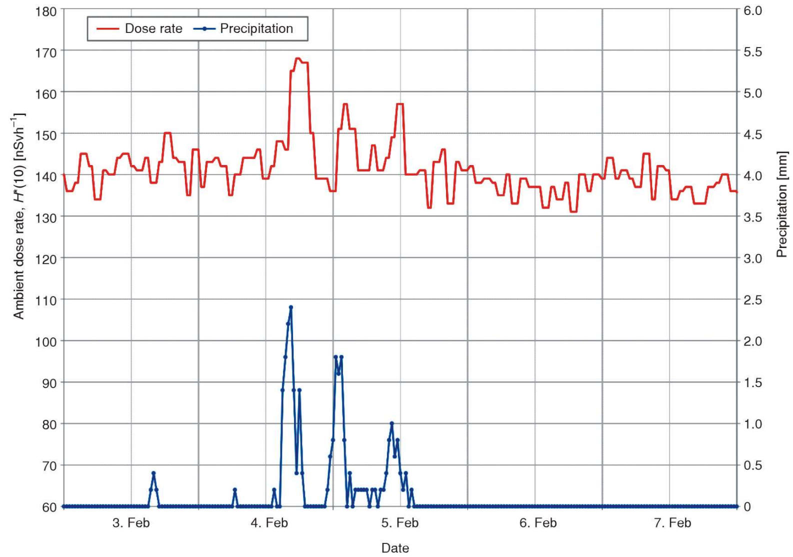

Figure 9. Ambient dose equivalent *H(10) rate and precipitation during February 2020, with special analysis of the rainiest period (03.02.2020-07.02.2020)**

INTERLABORATORY COMPARISON

Interlaboratory comparison is the organized performance and evaluation of the results of tests of the same parameters by two or more laboratories under predetermined conditions. These requirements are also described in the international standard ISO/IEC 17025:2017 [3].

As part of the interlaboratory comparison, the continuous measurement results of the ambient dose equivalent *H**(10) rate in the air of two laboratories were compared. Two participating laboratories were the Department for Radioactivity Examination and Environmental Protection in PC NFS and the Department of Radiation and Environmental Protection, Vinča Institute of Nuclear Sciences in Belgrade.

The PC NFS used the Multifunctional Gamma Radiation Monitor MFM203 with two GM probes (AMES Environmental Measurement Systems), and Vinča used the Stationary Alarm DMRZ-M15A Monitor with two plane-parallel pancake GM tubes inside the probe, with the measurement range from 50 $nSvh^{-1}$ to 1 $mSvh^{-1}$ [11]. Both probes are suitable for a photon energy range from 60 keV to 1.3 MeV.

Both dosimeters were calibrated in the Vinča Institute of Nuclear Sciences – Secondary Standard Dosimetry Laboratory according to International Organization for Standardization standard ISO 4037 [12-14].

The results of the interlaboratory comparison from 08.12.2021 to 13.12.2021 are shown in fig. 10. Additionally, a graphical representation of the measurement results during periods of elevated rainfall is included, highlighting a conspicuous correlation.

Due to the continuous nature of the measurements lasting 6 months, and large amounts of data, we could not present the entire data in this paper. That is why we did graphical representations of the dose rate during the significant rainy periods when the relation with the ambient dose equivalent rate in the air is particularly pronounced, as shown in fig. 10.

The organizer of the interlaboratory comparison statistically and graphically processed all the data. The

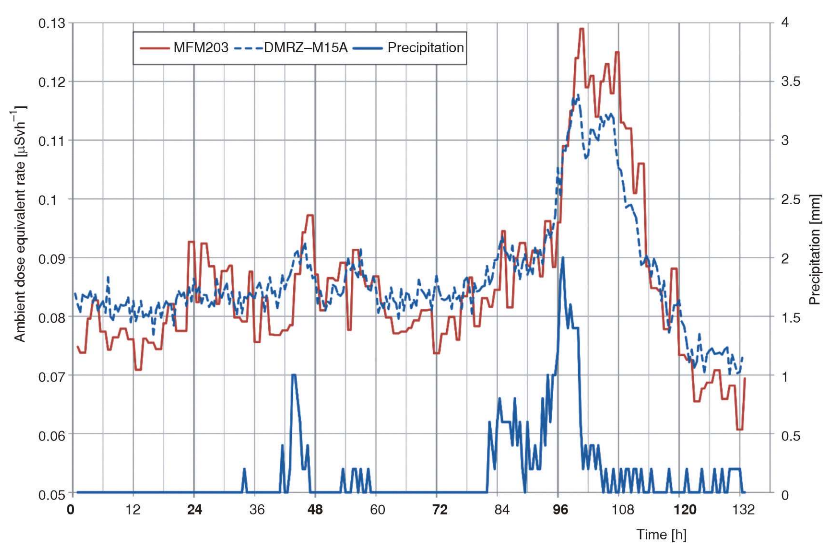

Figure 10. Ambient dose equivalent *H(10) rate and precipitation during inter-laboratory comparison for period 08.12.2021-13.12.2021**

graphical presentations in fig. 10 show a clear match between the measurement results. Both instruments also have a good response to precipitation and a strong relation between the ambient dose equivalent $H^*(10)$ rate and the amount of precipitation.

CONCLUSIONS

This paper presents the characteristics of the monitoring system for ambient dose equivalent $H^*(10)$ measurement and the automatic meteorological station used in the vicinity of nuclear facilities in the PC NFS. The measurement results of ambient dose equivalent and precipitation were gathered and analyzed.

Ambient dose rate diurnal variation caused by humidity changes increasing the dry deposition of radon progeny concentration was not observed due to the system not being sensitive enough for such small variations. However, the wet deposition of radon with progeny during the rainout was detected. A relationship between precipitation volume and dose rate increases was observed. An analysis of the dose rate increase and decrease in conjunction with meteorological data can help determine if the increase was caused by the radon washout or the presence of radioactive effluents from nuclear facilities, whether distant or *on-site*.

To ensure proper operation the system is regularly calibrated in an accredited laboratory. Also, a laboratory intercomparison was organized. The results were analyzed and are satisfactory, as the measured values of the difference between two systems at the same location were in the range of their measurement uncertainty. Statistical analysis of the intercomparison data showed a robust correlation between ambient dose equivalent $H^*(10)$ rate increases and precipitation levels.

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РЕЛАЦИЈА АМБИЈЕНТАЛНОГ ЕКВИВАЛЕНТА ЈАЧИНЕ ДОЗЕ И КОЛИЧИНЕ ПАДАВИНА ТОКОМ МОНИТОРИНГА РАДИОАКТИВНОСТИ У ОКОЛИНИ НУКЛЕАРНИХ ПОСТРОЈЕЊА

Део праћења радиоактивности у животној средини у близини нуклеарних постројења обухвата испитивање нивоа радиоактивне контаминације ваздуха, као и релевантна метеоролошка мерења на микролокацији, ради процене нивоа и контроле изложености становништва и животне средине. У раду је приказана корелација између промене јачине амбијенталног еквивалента дозе гама зрачења у ваздуху и количине падавина услед спирања атмосфере. Мерења су вршена на локацији метеоролошки стуб на 114 m надморске висине. Да би се приказао однос између измерених вредности амбијенталног еквивалента дозе и количине падавина, коришћена је варијација ових параметара током најкишовитијих месеци у 2019. и 2020. години. Међулабораторијско поређење је приказано да би се доказала валидација система. Анализа овог односа јасно показује утицај интензивне кише на јачину амбијенталног еквивалента дозе, што оправдава измерене краткотрајне повишене вредности које, у овом случају, нису настале услед евентуалног акцидентналног испуштања радиоактивних ефлуента услед рада нуклеарних постројења.

Кључне речи: мониторинг, животна средина, метеоролошки параметар, падавина, нуклеарно постројење