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AIR AND PRECIPITATION TESTING AS PART OF ENVIRONMENTAL RADIATION MONITORING IN THE VICINITY OF NUCLEAR FACILITIES

JOVANA KNEŽEVIĆ
PC „Nuclear Facilities of Serbia“, Mike Petrovića Alasa 12-14, Vinča, Belgrade, jovana.knezevic@nuklearniobjekti.rs

NATAŠA LAZAREVIĆ
PC „Nuclear Facilities of Serbia“, Mike Petrovića Alasa 12-14, Vinča, Belgrade, natasa.lazarevic@nuklearniobjekti.rs

SOFIJA FORKAPIĆ
Faculty of Sciences, University of Novi Sad, Trg Dositeja Obradovića 3, Novi Sad, Serbia, sofija@df.uns.ac.rs

KRISTINA BIKIT ŠREDER
Faculty of Sciences, University of Novi Sad, Trg Dositeja Obradovića 3, Novi Sad, Serbia, kristina.bikit@df.uns.ac.rs

VESNA RADUMILO
PC „Nuclear Facilities of Serbia“, Mike Petrovića Alasa 12-14, Vinča, Belgrade, vesna.radumilo@nuklearniobjekti.rs

DALIBOR ARBUTINA
PC „Nuclear Facilities of Serbia“, Mike Petrovića Alasa 12-14, Vinča, Belgrade, dalibor.arbutina@nuklearniobjekti.rs

Abstract: *Public company „Nuclear Facilities of Serbia“ is the only operator of nuclear facilities and holder of licenses to perform nuclear activities in the country. By the current legislation, environmental radiation monitoring in the vicinity of nuclear facilities is performed to assess the level and control the external exposure of the population and environment to ionizing radiation due to the operation of nuclear facilities. This paper presents an overview of the representative environmental radiation monitoring results regarding radioactive contamination level and ambient gamma dose rate equivalent in air and precipitation for the year 2021. The obtained results show that there is no adverse influence on the population and environment and the radiation risk for individuals in the population is negligible.*

Keywords: *monitoring, environment, radioactivity, testing, nuclear facility*

1. INTRODUCTION

Environmental radiation monitoring is a set of measurements, processing and interpretation of the results of measurements of radiation and meteorological parameters in order to assess the level and control the exposure of the population and the environment to ionizing radiation due to the operation of nuclear facilities. Systematic examination of radioactivity in the environment includes the following activities, according to the current legislation [1,2]:

- Measurements of radiation parameters, including:
  - Control of the level of radioactive contamination in the environmental samples (air, precipitation, surface water, river sediment, potable water, ground water, soil, food);
  - Examination the level of external radiation – measurements of ambient gamma dose equivalent and continuous measurements of ambient gamma dose rate equivalent);
- Measurements of meteorological parameters;
- Mathematical modeling of the distribution of radionuclides in the boundary layer of the atmosphere.

In accordance with the national legislation [2], Public company „Nuclear Facilites of Serbia“ (PC NFS) conducts measurements of radiation parameters by examining the level of external radiation, measuring meteorological parameters and mathematical modeling. An independent authorized legal entity, in accordance with the [2], conducts measurements of radiation parameters by controlling the level of radioactive contamination of the environment. Radioactive contamination measurements are performed by Department of Physics, Faculty of Science, University of Novi Sad, Serbia. The sampling locations are shown in table 1 [3].

Table 1. Annual program of examination of the level of radioactive contamination and the level of external radiation in the environment in the vicinity of nuclear facilities in PC NFS [3]

LOCATION	FREQUENCY	TYPE OF ANALYSIS
RADIOACTIVE CONTAMINATION LEVEL CONTROL IN AIR SAMPLES		
Reference point: 1. Green hill, Environmental Protection Agency *Three sites at the location of PC NFS with the most frequent winds, which are directed towards Vinča settlement:* 1. Weather station mounting pole 2. Radioactive waste (RAW) storage 3. Elementary school (in the vicinity of PC NFS)	Continuous collection, cumulative monthly sample, monthly measurement, 12 times a year	Gamma spectrometric analysis
Two sites at the location of PC NFS with the most frequent winds, which are directed towards Vinča settlement: 1. RAW storage 2. Elementary school (in the vicinity of PC NFS)	Continuous collection – cumulative quarterly sample, quarterly measurements, 4 times a year.	Determination of ^{90}Sr
RADIOACTIVE CONTAMINATION LEVEL CONTRON IN PRECIPITATION SAMPLES		
Reference point: 1. Green hill, Environmental Protection Agency *Two sites with the most frequent winds, which are directed towards Vinča settlement:* 1. Elementary school (in the vicinity of PC NFS) 2. Water station in Vinča settlement *Two sites at the location of PC NFS:* 1. Weather station mounting pole 2. „Vinča center“	Continuous collection, collected monthly sample, monthly measurement, 12 times a year	Gamma spectrometric analysis
Reference point: 1. Green hill, Environmental Protection Agency *One site with the most frequent winds, which are directed towards Vinča settlement:* 1. Water station in Vinča settlement *One site at the location of PC NFS:* 1. Weather station mounting pole	Continuous collection, collected monthly sample, monthly measurement, 12 times a year	Determination of ^{3}H
Reference point: 1. Green hill, Environmental Protection Agency	Continuous collection – cumulative quarterly sample, quarterly measurements, 4 times a year.	Determination of ^{90}Sr
AMBIENT GAMMA DOSE RATE EQUIVALENT IN AIR		
1. Research nuclear reactor RA (PC NFS) 2. Water station in Vinča settlement 3. Weather station mounting pole (PC NFS) 4. RAW storage (PC NFS) – two measurement points 5. Secure storage of high activity sources (PC NFS) 6. Waste processing facility (PC NFS)	Continuous automatic measurement	Measurement of ambient gamma dose rate equivalent

This paper presents the representative environmental radiation monitoring results regarding radioactive contamination level and ambient gamma dose rate equivalent, in air and precipitation samples, for the year 2021.

2. METHODS AND MATERIALS

In following chapters, methods for determing the activity concentration of radionuclides, as well as ambient gamma dose rate equivalent are described. All the testing methods for determing the presence of radionuclides in air and precipitation samples are accredited according to the standard SRPS ISO/IEC 17025:2017 [4].

2.1 Determination of radioactive contamination in the air and precipitation

Air sampling from the ground layer of the atmosphere was carried out by continuous suction using constant flow air sampling pumps (50-80 m^3/h). Monthly air samples collected on filter papers made of glass fibers (with pores of 1-2 μm) of high efficiency, by manufacturer F&J Specialty Products Inc., USA, were homogeneously packed in a cylindrical measuring vessel with dimensions 31 mm x 67 mm [5].

Precipitation samples were collected at the locations continuously with 25 m^2 surface samplers placed at a height of 0,8 m above the ground. Monthly precipitation samples were evaporated in dryers to a dry residue

according to the standard method [6].

Examination of gamma active radionuclides in air and precipitation samples was conducted by the gamma spectrometric method using High Purity Germanium (HPGe) detector according to [5]. Calibration of the HPGe detector was performed with certified reference material that contains mixture of gamma emitters in silica gel matrix of cylindrical geometry. Measurements of samples in contact geometry lasted for 80000 s. Detectors are connected to a digital spectroscopic processing using Canberra 1300 InSpector and multiport analyzer Canberra Multiport II. Gamma spectra were collected and analyzed using Canberra Genie 2000 Spectroscopy System software.

Examination of tritium ^{3}H in precipitation samples was performed in accordance with a validated method [7] on a liquid scintillation detector Quantulus 1220. This method involves mixing a distilled liquid sample with the scintillation cocktail in 8 ml : 12 ml ratio, in the original 20 ml measuring bottles. Quantulus 1220 has its own system of natural background radiation reduction consisting of active and passive protection. In order to determine „the window“ of ^{3}H in measured spectrum, the tritium standard with known activity concentration was measured as well as „background“ (distilled well water), and „the window“ was set to obtain the maximum figure of merit (FOM) factor. Preparation of the tritium standard, as well as „background“, was the same as preparation of the samples. Before the measurements were conducted, the samples were kept in a dark place for at least six hours to avoid the occurrence of luminescence.

Examination of strontium ^{90}Sr in air samples was performed as follows: the sample was dried at the temperature of 105°C. It was then homogenized with strong oxidizing agents and mineralized at the temperature of 600°C. The measured mass of mineral residue was dissolved in nitric acid with the addition of a known amount of inactive yttrium carrier. The separation of ^{90}Y from ^{90}Sr is based on a multi-stage extraction procedure with tri-butyl-phosphate, nitric acid and deionized water. The separated ^{90}Y was deposited as Y-hydroxide, which was transformed into yttrium oxide by annealing at the temperature of 900°C. The chemical yield of yttrium was calculated from the mass of obtained Y_2O_3. The activity concentration was determined by measuring the activity concentration of the separated yttrium in an anti-coincidence counter of low beta activities „INC-Instruments“ USA.

Examination of ^{90}Sr in water samples was conducted by a validated method based on the detection of Cherenkov radiation on a liquid scintillation counter. Radioactive strontium ^{90}Sr decays by beta decay into its progeny yttrium ^{90}Y, emitting beta particles with maximum energy E_{max} = 546 keV. ^{90}Y is also beta emitter with maximum energy of emitted beta particle E_{max} = 2280 keV, and the half-life value of ^{90}Y is 64 h. Due to its short half-life, ^{90}Y is in radioactive equilibrium with the parent nucleus ^{90}Sr. In this way, by measuring the activity concentration of ^{90}Y in the sample, the activity concentration of ^{90}Sr in the sample is estimated. Fast electrons that are emitted by the decay of ^{90}Y produce the Cherenkov radiation in water. Minimum energy that fast electrons need to have to produce the Cherenkov effect in water is 256 keV. The Cherenkov radiation belongs to visible and UV part of electromagnetic spectrum, so it can be identified by photomultiplier on a liquid scintillation detector. For detector calibration, a standard radioactive material (water solution $^{90}Sr/^{90}Y$) was used. The measurements were conducted on liquid scintillation detector Wallac 1220 Quantulus, that has high stability, good separation of alpha and beta signals based on the classification of pulse amplitudes based on their shape, passive protection made of lead and active protection against cosmic and other environmental radiation. Spectrum analysis was performed using Wallac WinQ 1220-307 Windows software, and for the data processing Wallac Easy View 1224-534 Spectrum analysis program.

2.2. Continuous measurements of ambient gamma dose rate equivalent

Continuous measurements of ambient gamma dose rate equivalent in the air were carried out to estimate the radiation field variations in the vicinity of nuclear facilities, as well as early warning of accidents. The ambient gamma dose rate equivalent in the environment generally depends on the geographical location as well as on the radioactive content of the local rocks and soil. In addition, these values also depend to a certain extent on different meteorological conditions (precipitation, humidity, wind, etc.). During the rainy season, there is a washing of radioactive elements from the atmosphere to the ground and an increase in the ambient radiation level in a short period of time. Therefore, there is justified to examine and determine the correlation between radiation and meteorological parameters, since from the point of view of radiation safety and radiation protection, it is very important to evaluate the influence of meteorological parameters and phenomena that can lead to changes in the values of radiation parameters, in order to unambiguously determine when detecting increased ambient gamma dose rate equivalent in the air, whether it is a potential radiation/nuclear accident, or an increase in the consequences of meteorological conditions in the environment at a given time.

Concentration of pollutants from artificial sources in the environment, in general, is monitored by a monitoring network with a limited number of discrete points. These measurements were performed with multifunctional gamma monitors. They are portable instruments, designed to automatically perform all necessary functions as independent local monitors. One monitor is connected to two energy-compensated Geiger-Miller counters (probes), with different sensitivities that can cover the range from natural background radiation to accidental levels (from 50 nSv/h to 1 Sv/h). These measurements were carried out according to the standard [8].

The data were collected continuously, during 24 hours and sent in half-hourly intervals to the database on the central computer in the control room where data acquisition, processing, storage, presentation and interpretation of obtained values were carried out. The

entire system is connected to the eEMIS application. With the help of the eEMIS application, data processing for the needs of operational tasks as well as for research purposes are performed [9].

The amount of precipitation, for the purpose of the correlation, was measured by an electric rain gauge with a heater, placed at a height of 1 m above the ground. Precipitation data is displayed on the central computer in the control room.

3. RESULTS AND DISCUSSION

3.1. Radioactive contamination in the air and precipitation

Figures 1 and 2 show the values of activity concentration of ^{7}Be and ^{210}Pb in air, respectively. Activity concentration of ^{137}Cs in air is below the value of minimum detectable activity (MDA), thus not presented as figure. Figure 3 shows the cumulative quarterly measurements of activity concentration of ^{90}Sr.

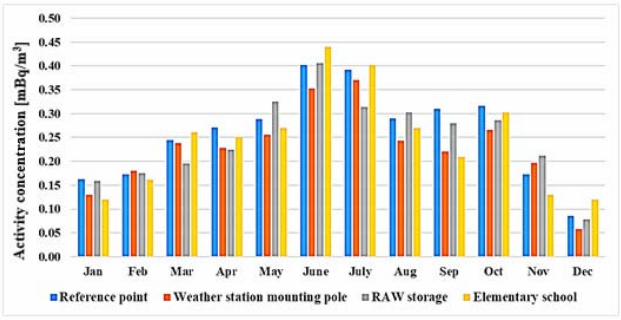

Figure 1. Activity concentration of ^{7}Be in air in mBq/m^3 for different locations during the year of 2021

It is known that production of cosmogenic radionuclide ^{7}Be depends on interaction between cosmic rays and upper layers of atmosphere. The production rate is varied by the solar modulation of galactic cosmic rays which is controlled by the solar magnetic field and, in turn, by solar activity [10], hence the seasonal variations of ^{7}Be seen on figure 1. The highest values of activity concentration are in June and July, while lower values were obtained during winter months.

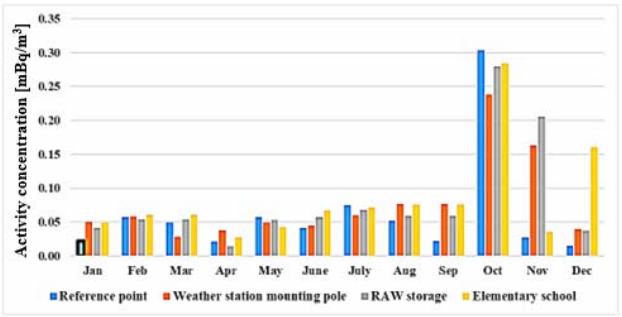

Figure 2. Activity concentration of ^{210}Pb in air in mBq/m^3 for different locations during the year of 2021. The framed bar (▭) represents MDA, since the value is below MDA

The observed increase in the activity concentration of ^{210}Pb in air in October 2021 is not due to operation of nuclear facilities, since the increase was also detected at the reference point with the highest value of activity concentration among all measurement points. The obtained value is in accordance with the results of environmental radioactivity monitoring in Serbia, where the activity concentration for the same measurement point is in interval 0,3 – 1,5 mBq/m^3 [11].

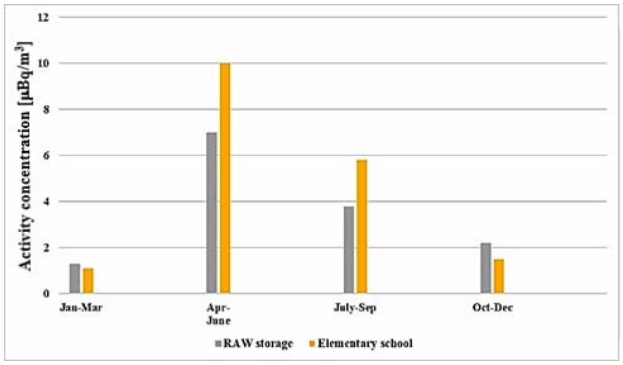

Figure 3. Activity concentration of ^{90}Sr in air in μBq/m^3 for different locations during the four quartals of 2021

Figures 4, 5, 6 and 7 show the values of activity concentration of ^{226}Ra, ^{232}Th, ^{40}K and ^{238}U in precipitation samples, respectively. Activity concentration of ^{235}U, ^{137}Cs and ^{3}H in precipitation is mostly below MDA, thus not presented as figure. The cumulative quarterly values of activity concentration of ^{90}Sr in precipitation are below MDA.

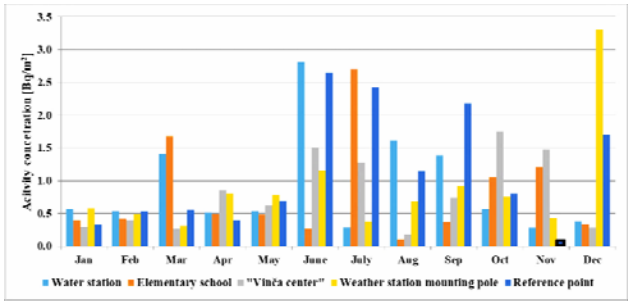

Figure 4. Activity concentration of ^{226}Ra in precipitation in Bq/m^2 for different locations during the year of 2021. The framed bar (▭) represents MDA, since the value is below MDA

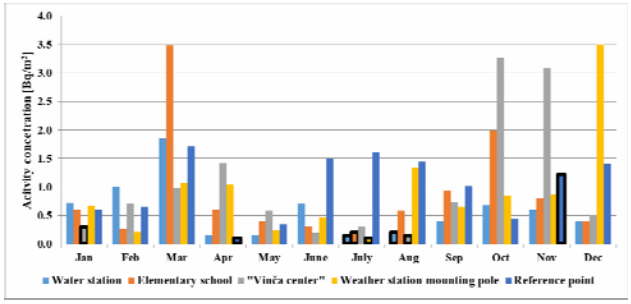

Figure 5. Activity concentration of ^{232}Th in precipitation in Bq/m^2 for different locations during the year of 2021. The framed bars (▭) represent MDA, since these values are below MDA

It can be seen on figures 4 and 5 that values of activity

concentration of ^{226}Ra and ^{232}Th differ from one location to another. The existence of correlation between detected higher values and the position of the locations cannot be established, given the amount of data and parameters that influence the results (eg. geographic area/terrain, meteorological conditions of the location). Activity concentration of both radionuclides fluctuates in interval that is consistent with characteristic data for precipitation.

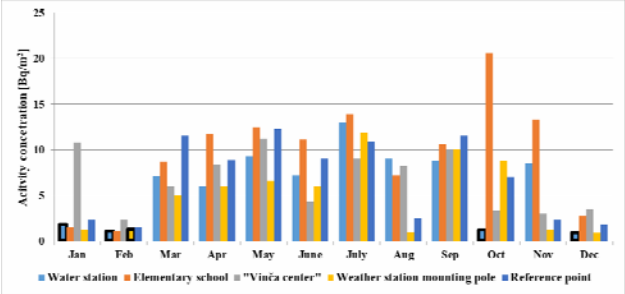

Figure 6. Activity concentration of ^{40}K in precipitation in Bq/m^2 for different locations during the year of 2021. The framed bars (▫) represent MDA, since these values are below MDA

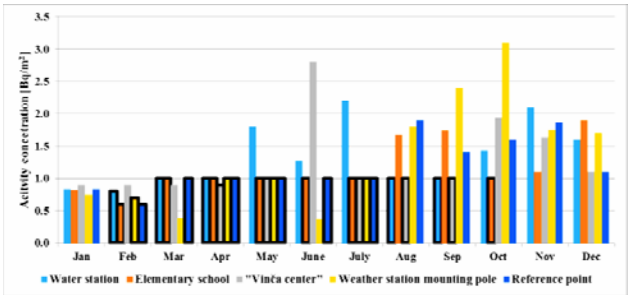

Figure 7. Activity concentration of ^{238}U in precipitation in Bq/m^2 for different locations during the year of 2021. The framed bars (▫) represent MDA, since these values are below MDA

The results for radionuclides ^{40}K and ^{238}U shown in figures 6 and 7, respectively, vary within the normal range for environmental samples.

3.2. Measurements of ambient gamma dose rate equivalent in the air

Figure 8 shows the monthly average values of ambient gamma dose rate equivalent in nSv/h for the measurement points according to table 1 in the vicinity of nuclear facilities. Figure 9 presents monthly variations of the daily mean values of ambient gamma dose rate equivalent for all six measurement points.

The obtained values of the ambient gamma dose rate equivalent in the air did not show any deviations from the mean values obtained by multi-year measurements on this site. The local maxima of ambient gamma dose rate equivalent in the air, which can be seen on the figure 9, are the result of intense precipitation with which there is a strong correlation. The characteristics of the measuring devices are such that even the slightest change of ambient gamma dose rate equivalent is registered due to washing of radioactive elements from the atmosphere during precipitations. Figure 10 represents correlation between ambient gamma dose rate equivalent [nSv/h] and the amount of precipitation [mm].

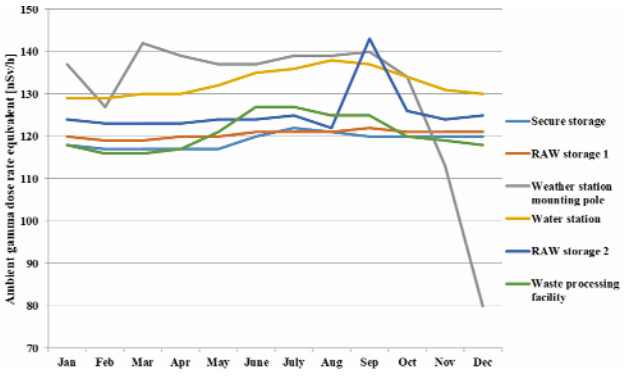

Figure 8. Monthly average ambient gamma dose rate eqivalent in the air [nSv/h] for different locations for the year 2021

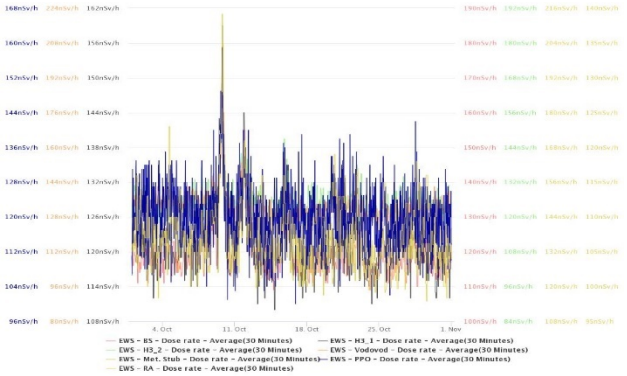

Figure 9. Monthly variation of the daily mean values of ambient gamma dose rate equivalent [nSv/h] at the PC NFS, October 2021

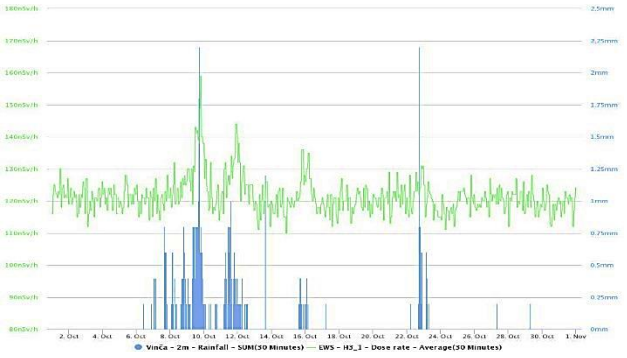

Figure 10. Correlation between ambient gamma dose rate equivalent [nSv/h] and precipitation [mm] for „Weather station mounting pole“ at PC NFS, October 2021

4. CONCLUSION

The presence of radionuclides of natural origin was detected in the samples, in activity concentration intervals that are characteristic for the investigated types of samples from the environment.

The activity concentration of artificial radionuclide ^{137}Cs is below MDA in both air and precipitation samples, for the examined period.

In precipitation samples, the acitivty concentration of radionuclide ^{3}H is below MDA, for the examined period.

The activity concentration of ^{90}Sr in air samples, for all four quarters of 2021, range from 1 μBq/m^3 to 10 μBq/m^3. The activity concentration of the produced radionuclide ^{90}Sr in precipitation is below MDA in all samples.

The values of ambient gamma dose rate equivalent in the air samples in the vicinity of nuclear facilities during 2021 are within the natural background radiation variation range. The increase of the values due to the atmospheric factors does not exceed the double value of natural background radiation (200 nSv/h).

The obtained values of activity concentration of radionuclides and ambient gamma dose rate equivalent show that the operation of nuclear facilities did not contribute to an increase in the level of exposure of the population and the radiation risk for individuals in the population is negligible. Along with this comes the fact that research nuclear reactor RA is out of operation mode for 30 years, fresh and spent nuclear fuel were repackaged and transported to the country of origin, the waste processing facility has not yet started its trial run, and both licenced RAW storage and secure storage for high activity sources are operating in safe and secure manner.

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