

## Research Article

# State of the surface waters in the Mesta River basin, after the reclamation of the Eleshnitsa uranium deposit

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## Abstract

In the period from 1955 to 1992, uranium ore was mined and processed in the Mesta River basin around the village of Eleshnitsa, region of Razlog. The ore processing plant was built and a tailings storage facility was built next to it. In the period 2002–2006, the tailings storage was reclaimed, and in 2011–2012 the ore mines were also reclaimed. For the treatment of drainage water, a treatment plant was built in the valley of Valchoto Dere River, a left tributary of the Mesta River. The radiological monitoring carried out by the Basin Directorate "West Aegean Sea Region" reveals a generally good quality of the surface waters in terms of the content of uranium and radium (<sup>226</sup>Ra) outside the area of the former uranium mining and the tailings storage facility. In this area, in some years a high content of natural uranium was found, with concentrations from 0.54 to 67.40 mg/l on average per year. The values significantly exceed (by 1.8 to 224.6 times) the norm regulated in the regulation for radiation protection and safety from the liquidation of the consequences of uranium mining. The spread of uranium is limited to a small area shortly downstream from the sources of contamination, but the potential risk to the local ecosystem should not be neglected. Radium, in contrast to uranium, has concentrations below the permissible limits in the twelve years studied. Its content varied from 0.025 to 0.11 Bq/l on average annually. The results show that the Mesta River near the border with the Republic of Greece is not contaminated with the studied radionuclides.

**Key words:** Balkan peninsula, environmental quality, pollution, radiological monitoring



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## 1. Introduction

Uranium mining is controversial mainly because of the associated environmental and human health impacts. Globally, it has generated  $938 \times 10^6$  m<sup>3</sup> of waste (Abdelouas 2006). The radioactivity of this waste depends on the quality of the mined ore and varies from less than 1 Bq/g to more than 100 Bq/g. Waste from the uranium industry is often associated with increased concentrations of highly toxic radionuclides, which are a major source of surface and groundwater pollution (Abdelouas 2006; Fritz and Arntzen 2007). This problematic issue is also advocated in the research of Beneš (1999); Dewar (2019); Srivastava et al. (2020); Horai et al. (2022).

The uranium mining industry in Bulgaria developed intensively in the second half of the 20<sup>th</sup> century. In the period 1946–1990 about 80 uranium deposits

were explored, and 47 out of them were developed (Petrov and Bozhkov 1987). One of them is the deposit near the village of Eleshnitsa—the region of Razlog in South-West Bulgaria. Uranium mining was carried out in it from 1955 to 1992, when by a decree of the Council of Ministers, uranium mining in Bulgaria was stopped.

Subsequently, the cessation of this sector of the national economy did not solve the emerging environmental problems. The closed uranium mines, old geotechnical installations and facilities, the tailings ponds, as well as the polluted production sites represent one very significant source of contamination, which will continue to exert a negative impact in the coming decades along with other mining-caused pollutions (Cholakova 2019; Penin and Zhelev 2020; Kotsev and Stoyanova 2022). In recent years, studies of environmental hazards that come from abandoned uranium mines in specific regions of the Balkan Peninsula have been carried out in Bulgaria (Bogoev et al. 2010; Tsekova and Bogoev 2010; Petrova 2012; Dikov and Bozhkov 2014; Ivanova et al. 2015; Tsekova and Lozev 2017), in Serbia (Dragović et al. 2014; Tanić et al. 2014), in Turkey (Turhan et al. 2012), in Croatia (Šoštarić et al. 2017; Petrinc et al. 2018; Ivanić et al. 2019) and other countries. Research related to the impact of the anthropogenic factor on the surface and underground water in different parts of Bulgaria was conducted by Marcheva et al. (2023) and Stefanov et al. (2023).

After the cessation of uranium mining and the liquidation of the mines, the uncontrollable leakage of mine waters continues for a long time due to the lack of facilities for its control and management (Bedrinov 2000). These mine waters, which are enriched with radionuclides, are transported to the river system of the Mesta River, and thus the pollution spreads over large areas.

To clarify the environmental situation regarding the pollution of surface and underground waters during and after the cessation of uranium mining, a monitoring network was built in the basin of the Mesta River. In several studies by Nikolova (2020), Stoyanov et al. (2021), and Kolev et al. (2014) the data from these observations were analyzed and it was established that during the active mining activity, the surface and underground waters were mainly contaminated with radium ( $^{226}\text{Ra}$ ) and less with uranium.

One of the main unsolved problems related to the uranium mining conducted in the past has been the management and complex purification of the radionuclide-contaminated water outflow from the “Eleshnitsa” uranium deposit. This represents a serious radio-ecological risk and a potential danger to human health.

Therefore, the present study aims to assess the current ecological state of surface waters in terms of the content of the radionuclides uranium and radium ( $^{226}\text{Ra}$ ), which was established after the liquidation and reclamation of the production facilities and the tailings storage facilities in the Eleshnitsa deposit.

## 2. Material and methods

### 2.1. Study area

The mining of uranium ore from the Eleshnitsa ore deposit is carried out through 5 shafts, 84 adits, 4 quarries, and one acid block underground leaching section. 35 pit dumps were built. When making the adits, numerous pit dumps

were formed (~ 60 places), covering a total area of 147,270 m<sup>2</sup> and with a total volume of 2,435,240 m<sup>3</sup> (Nikolova 2000).

The ore processing plant “Zvezda” (“Star”), the second largest in Bulgaria, was built in 1965, and since 1969, the tailings storage facility has been in operation. Currently, the tailings storage facility contains about 9 million tons of hydrometallurgical wastes, including around 730 tons of uranium. In 1995, it was rehabilitated, and in the period 2002–2005, a complete reclamation was carried out. To treat the drainage water from the tailings, a treatment plant was built in the valley of Valchoto Dere River.

The study area includes the catchments of the Zlataritsa River and Valchoto Dere River, which drain the deposit and are part of the Mesta River basin. A network for radiological monitoring of the Basin Directorate of the West Aegean Sea Region was built on them, the data of which are analyzed in the present study (Fig. 1).

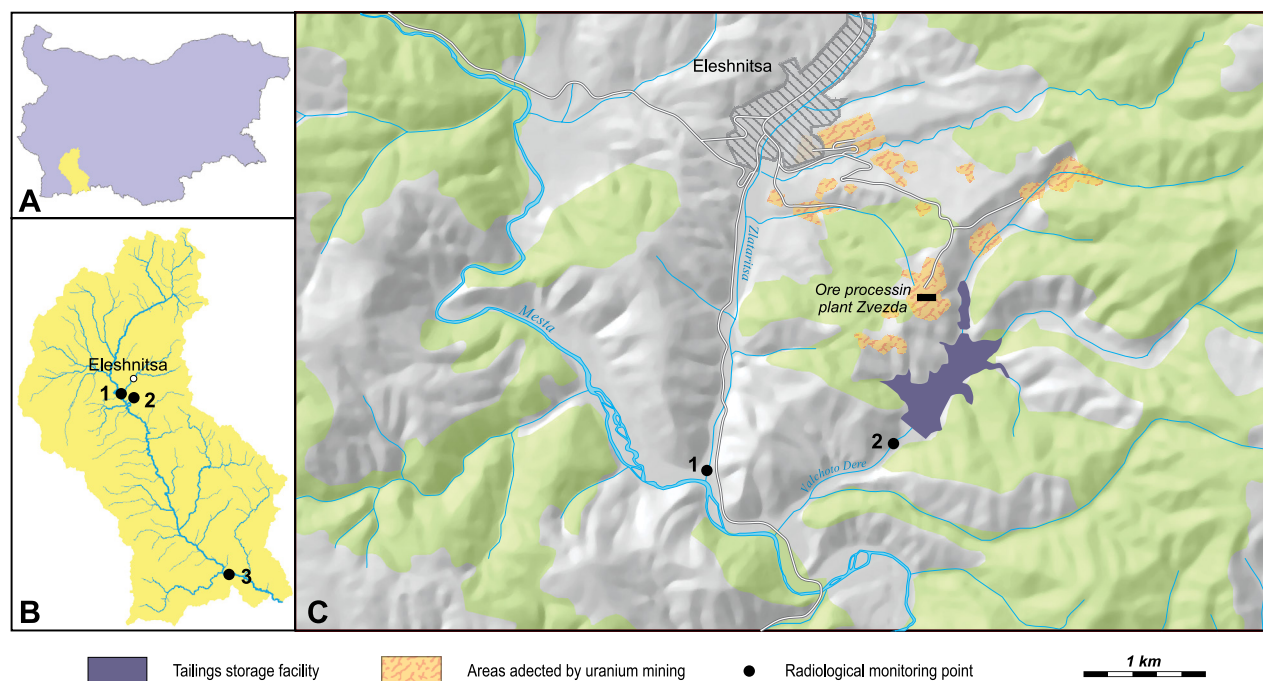


Figure 1. A map of the research area and the monitoring points.

The first radiological monitoring point is located at the mouth of the Zlataritsa River. River length is 21.3 km and the area of its catchment area is 111.2 km<sup>2</sup>. The river does not dry up and does not freeze in winter, because waters from warm mineral springs pour into it. It flows through the village of Eleshnitsa and in its lower course below the village it is used to irrigate fields and vegetable gardens. The main part of the mining operations of the “Eleshnitsa” uranium deposit was concentrated in the Zlataritsa river basin—the mine galleries (shafts), soles and quarries, the places of soda and heap extraction, sorption, and geotechnology. Through the surface and subsurface runoff, these excavation sites are drained by the Zlataritsa River. The second point for radiological monitoring is located under the wall of the tailings storage facility in the basin of the Valchoto Dere River, a left tributary of the Mesta River. The basin of the

Valchoto Dere River is significantly smaller in area than that of Zlataritsa River—about 16 km<sup>2</sup> and a little over 6 km long. On the site where the tailings storage facility is located, two ravines merge—the Dindirishko ravine from the northeast and the Oreovsko ravine from the southeast, which feed the area with surface flowing water. A system of concrete collectors has been built in these ravines to divert the waters from them and take the waters to the Valchoto Dere River under the wall.

The third point for radiological monitoring is located on the Mesta River downstream from the town of Hadzhidimovo and from the confluence of the Matnitsa River (Fig. 1). The cross-border Mesta River is 273 km long, 126 km of which are on Bulgarian territory. The river flows through Southwestern Bulgaria in an area recognized as the country's most abundant water supply area. Within the country, the river catchment has an area of 2,767 km<sup>2</sup> and has the highest average altitude among Bulgaria's main rivers. According to data from the National Institute of Meteorology and Hydrology, the average annual runoff for the period 1993–2008 was 18.9 m<sup>3</sup>/sec at Hadzhidimovo station (National Institute of Meteorology and Hydrology). Mesta River waters are used for irrigation in the middle and lower catchment sections.

## 2.2. Data and methods

In the three mentioned points, radiological monitoring is carried out four times a year. The samples were collected at a minimum of 30 cm above the river bottom and at the same depth from the water surface. On sites where discharge was smaller, the sampling was done at an average height between the water surface and the river bottom. Sampling was done during all seasons of the year, on sites that were far enough from the river bank and roads, to avoid admixture with salt, sand, and other macro and micro elements from road surfaces and products from riverbank erosion. Samples were collected directly into laboratory bottles with a capacity of 3 l. Containers were submerged in the homogenous zone of the river, to not include the surface water film or water whirls. Collected samples were stabilized and stored according to Standard ISO 5667-3 and were subjected to analysis in a certified radiological laboratory.

The West Aegean River Basin Directorate is in charge of performing radiological monitoring of surface waters according to Bulgarian State Standard (BDS) EN ISO 5667-6: 2014/A 11:2020, which is coordinated with the European Standard ICS:13.060.10 13.060.45. The research period for two of the points is 12 years (from 2011 to 2022) and for the point at Valchoto Dere River—8 years (from 2012 to 2019).

The samples were analyzed in the radiological laboratory of the Environmental Executive Agency, under the Ministry of Environment and Water. The determination of the studied radionuclides is carried out by the Inductively coupled plasma mass spectrometry (ICP-MS) method, using the BSS EN ISO 17294 and BSS EN ISO 17294 -2 standards for uranium, and the BSS 12575:1975 standard for radium.

To compare the data, the indicators for the content of total uranium and radium from Ordinance № H-4/14.09.2012 for the characterization of surface waters were used (Ministry of Environment and Water 2012). In this Ordinance,

the content of total uranium is regulated at 0.04 mg/l (40 µg/l), and of radium ( $^{226}\text{Ra}$ ) at 0.1 Bq/l.

Ordinance № 1/11.1999 for radiation protection and safety during liquidation of the consequences of the uranium mining industry in Bulgaria (CPUAE et al. 1999) applies to rivers near uranium mining sites. The content of natural uranium here is 0.3 mg/l, and the specific activity of radium is 500 mBq/l (0.05 Bq/l).

### 3. Results and discussion

The data from the monitoring of the Basin Directorate of the West Aegean Sea region show that in the Zlataritsa River (monitoring point 1, Fig. 2) the uranium content varies on average between 0.000043 and 0.1147 mg/l. These concentrations do not exceed the norm regulated in Ordinance № 1 of 1999 for rivers near uranium sites. However, in eight of the twelve years studied, the uranium content in the waters was higher than the maximum permissible concentrations regulated in Ordinance № H-4 of 2012. The excesses were particularly high in 2017—3 times, and in 2020—2.7 times. This trend is due to the fact that with the liquidation of the technical facilities, the drainage pumps were turned off and in several mine galleries the level of underground water rose, so in some places, there was a leakage of mine water from boreholes and mine workings that had an outlet to the earth's surface. Mine waters in contact with mined and abandoned uranium ore in the galleries, extract and export part of the uranium to the surface waters. A study by Kolev et al. (2018) found that the prevailing flow rates of these mine waters were below 0.1 l/s, and uranium concentrations were low—below 0.04 mg/l. Their influence on surface water is minimal and has an extremely local character.

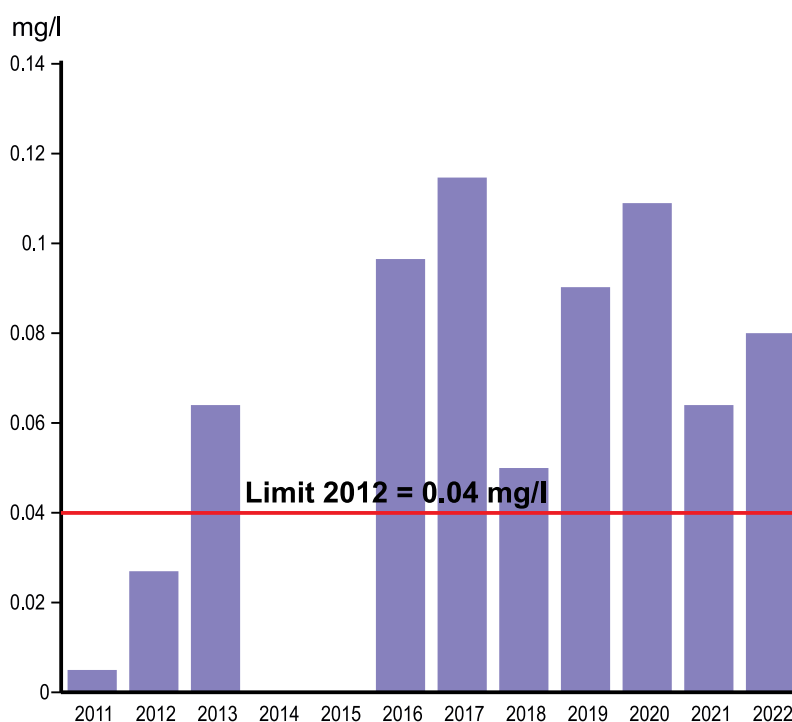


Figure 2. Annual uranium content in the waters of the Zlataritsa River.

For the twelve-year research period, the content of radium ( $^{226}\text{Ra}$ ) of the waters of the Zlataritsa River has varied from 0.025 to 0.043 Bq/l (Fig. 3). The variations are small and the concentrations are significantly below the permissible values of both regulations, approaching the Clarke values according to Ilyina et al. (1990).

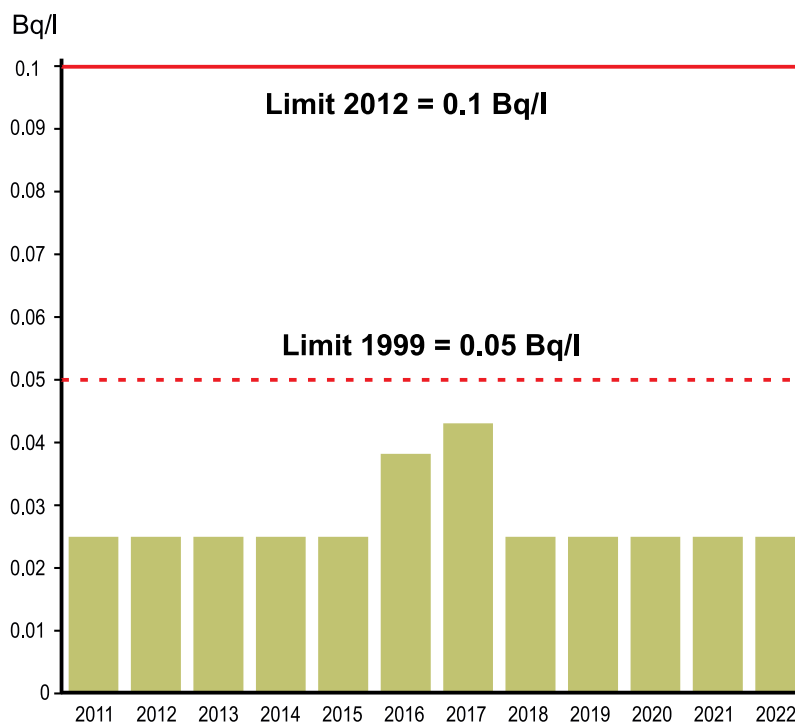


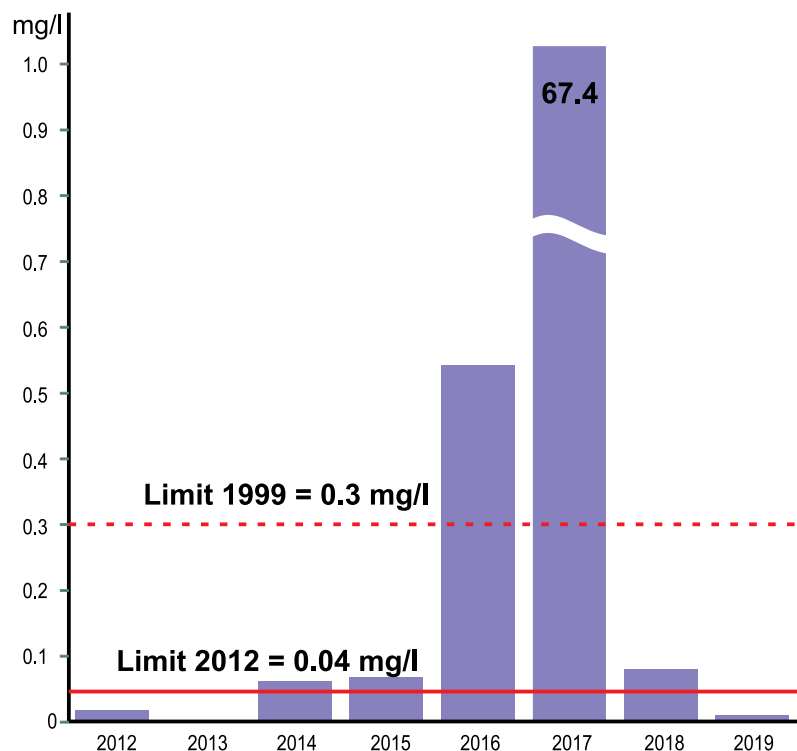
Figure 3. Annual content of radium ( $^{226}\text{Ra}$ ) in the waters of the Zlataritsa River.

According to previous research by Nikolova (2020), it was radium that was the main pollutant of surface waters during the period of uranium mining. This was due to the process of leaching the ore with sulfuric acid, a process that extracted the uranium and left the radium in the waste water. Now this activity has been stopped and a higher content of uranium and a significantly lower amount of radium ( $^{226}\text{Ra}$ ) is observed in the waters of Zlataritsa River. Due to the fact that it is no longer extracted, the uranium remains in the strata and galleries, shafts and adits, and the groundwater brings it to the surface and into the waters of the larger rivers, the Zlataritsa River and the Mesta River.

In the waters of Valchoto Dere River—monitoring point 2, the main pollutant is once again uranium. The source of these high concentrations is the tailings storage facility (Fig. 1). Its excesses in 2017 were particularly high, reaching 1,685 times the maximum permissible concentrations (MPCs) for surface waters regulated in the 2012 regulation (Fig. 4). For the same year, uranium concentration exceeded the regulated values from 1999 by a magnitude of 225. These extremely high uranium contents were most likely due to the fact that 2017 was very rainy, with widespread floods throughout the country, and the heavy rainfalls infiltrated through the sealed cover of the tailings storage facility, which bottom is not well insulated. As a result, large amounts of radioactively contaminated water penetrated deep into the ground. Besides that, the tunnel under the tailings is not well reinforced and favours the flow of polluted



water under the wall, into Valcheto Dere river. The leakage of drainage water continues to this day, despite the rehabilitation and reclamation that have been carried out in order to capture and purify the waters in the constructed treatment plant.



**Figure 4.** Annual uranium content in the waters of Valchoto Dere River.

During the studied period, radium did almost not exceed the norm for surface water, except for 2017, when the values were 1.1 times above the MPC concerning Ordinance 4 of 2012 (Fig. 5). In most years from the studied period, its concentrations were higher compared to those from point 1—the mouth of the Zlataritsa River.

Kolev et al. (2014) and Kolev (2017) also found that the tailings storage facility has been a major emitter of radioactive substances and contaminates the underground water in the Valchoto Dere area even since the construction of the treatment plant. According to Mihailova et al. (2023), the deposit around the village of Eleshnitsa is the largest ore deposit in the valley of the Mesta River, exploited for a long time, and it is very difficult to separate the anthropogenic load with radionuclides derived from natural background.

Moving away from the mouth of the Zlataritsa and Valchoto Dere Rivers, the anthropogenic nature of the migrating radionuclides weakens.

This decrease is particularly noticeable at monitoring point 3 of the Mesta River after the town of Hadzhidimovo, a few kilometers upstream from the border with the Republic of Greece, where a significant drop is observed for uranium, and radium maintains its low values (Figs 6–7).

The content of both radionuclides is far below the norm regulated in Ordinance H-4 of 2012 for surface waters, approaching the Clarke concentrations in the river waters on land according to Ilyina et al. (1990). A prerequisite for the

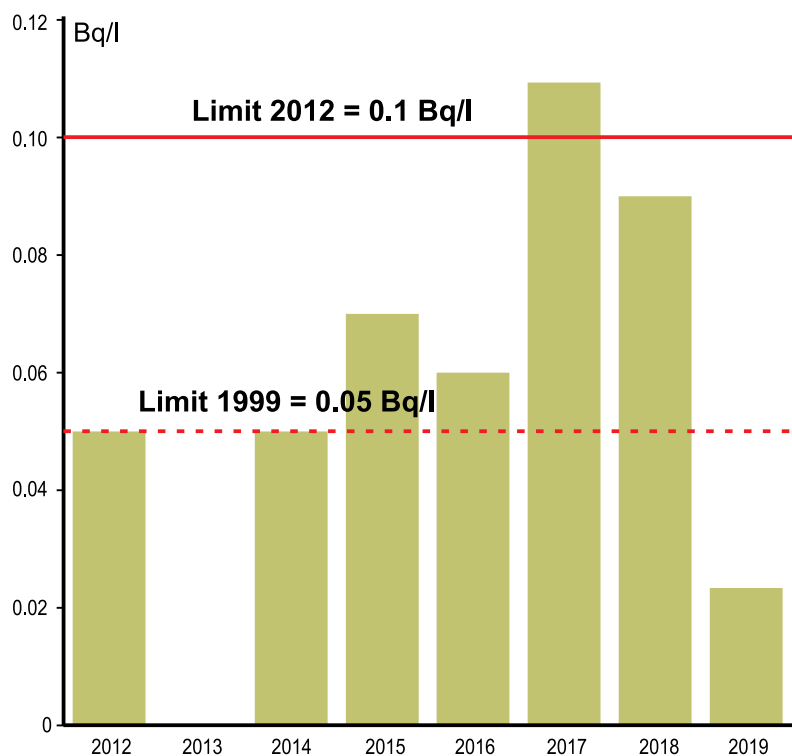


Figure 5. Annual radium content ( $^{226}\text{Ra}$ ) in the waters of the Valchoto Dere River.

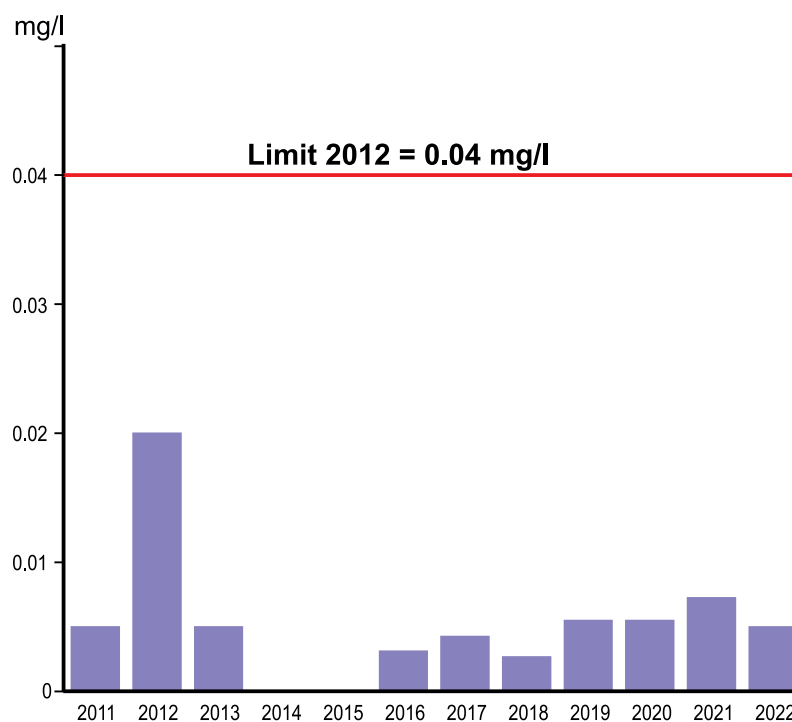


Figure 6. Annual uranium content in the waters of the Mesta River.

low values is the considerable distance from the emitters, as well as the great dilution of the pollutants in the abundant runoff of the Mesta River. Additionally, these low levels could be also explained by the presence of a geochemical barrier in the form of bottom sediments along the Mesta River, which sediments are active sorbents of radionuclides.



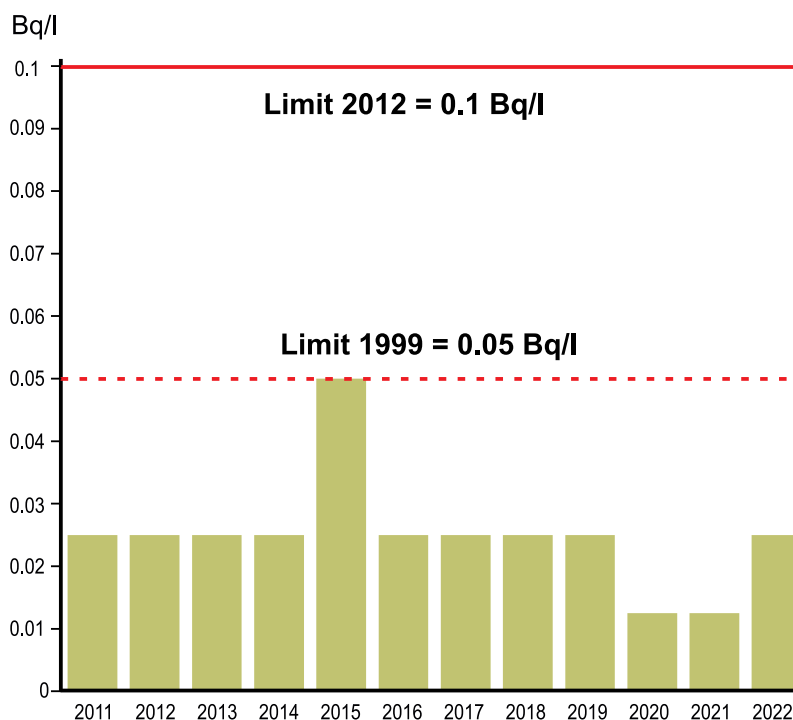


Figure 7. Annual radium content ( $^{226}\text{Ra}$ ) in the waters of the Mesta River.

#### 4. Conclusion

The results of the studies have proved that the area of Eleshnitsa's former uranium mine deposit remains a local source of pollution. Regardless of the activities carried out and the resources invested in eradicating the effects of uranium mining, the leakage of uranium and its associated elements into surface waters has continued for almost twenty years afterward.

Particularly affected by the uranium mining industry are the water courses of the Valcheto Dere River and Zlataritsa River, which drain the deposit and the tailings storage facility. In certain rainy years, the concentrations of uranium in these rivers exceeded many times the precautionary standards.

Moving away from the source of pollution, towards the border with the Republic of Greece, there is a significant dilution of the studied radionuclides as a result of the long journey and the self-purifying function of the Mesta River, which has more water. However, this does not mean safety, because polluted waters have been flowing untreated into the Mesta River for more than 60 years, and the studied radionuclides, according to the peculiarities of their migration, were gradually deposited in the bottom sediments downstream.

These deposits represent a kind of depot for the accumulation of radionuclides, and when geochemical conditions change or very high waters and floodings occur, they can become a source of pollution for the Mesta River. For a more complete clarification of this process, it is necessary to conduct new studies. Additional results are needed to better predict the behavior of pollutants in the waters of the studied area.

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## Additional information

### Conflict of interest

No conflict of interest was declared.

### Ethical statement

No ethical statement was reported.

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### Author contributions

All authors have contributed equally.

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### Data availability

All of the data that support the findings of this study are available in the main text or Supplementary Information.