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RADIONUCLIDE CONTAMINATION IN AGRICULTURAL AND URBAN ECOSYSTEMS: A STUDY OF SOIL, PLANT, AND MILK SAMPLES

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ABSTRACT. This study investigates the presence of radionuclides in soil, plant, and milk samples from rural and urban areas. Using gamma spectrometry, the activity concentrations of radionuclides were measured to assess potential exposure pathways for external gamma radiation and radionuclide transfer through the food chain. Higher activity levels were generally observed in rural environments, while certain plant and milk samples revealed trace contamination. These findings underscore the importance of continuous environmental monitoring to safeguard food safety and mitigate risks associated with radiological contamination.

Keywords: radionuclide contamination, soil, plant, milk.

INTRODUCTION

Radiation exposure remains a persistent concern for human health, with sources ranging from natural occurrences to anthropogenic activities. Understanding the distribution and levels of radioactivity in the environment is crucial for assessing potential risks to populations (UNSCEAR, 2008; MONGED *et al*., 2020). In this context, soil serves as a reservoir

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for various radionuclides, acting as a pathway for their transfer to plants and, subsequently, to the food chain (KEUM *et al*., 2007). Moreover, soil radioactivity contributes significantly to external gamma radiation exposure. The analysis of milk samples is of great importance for assessing the level of radionuclide contamination in this key food product (GIRI *et al*., 2011; CAMPBELL *et al*., 1961). Prior to sampling, rigorous precautionary measures must be taken to ensure the integrity and representativeness of the collected samples. Prolonged exposure to radionuclides through contaminated soil, plants, and milk can lead to significant public health risks, including increased cancer incidence and genetic damage. The ingestion of radionuclides, such as ^{137}Cs and ^{226}Ra, has been associated with internal radiation exposure, which accumulates over time and can affect vital organs. These potential health effects underscore the importance of evaluating radioactivity levels in environmental and food samples. Prior to sampling, rigorous precautionary measures must be taken to ensure the integrity and representativeness of the collected samples.

The city of Kragujevac, situated in Serbia, represents a dynamic urban environment where the interplay between natural and artificial sources of radiation merits investigation. Natural sources, including cosmic radiation and radionuclides inherent in the Earth's crust, intertwine with anthropogenic contributions such as nuclear accidents and atmospheric testing fallout. As a result of the anthropogenic activity of one of the most important artificial radionuclides, ^{137}Cs, with a half-life of 30.1 years, has a strong affinity for binding to the soil and is therefore a permanent environmental pollutant (DJELIC *et al*., 2016; ZHU *et al*., 2000; STÄGER *et al*., 2023).

This study endeavors to elucidate the radioactivity landscape in the vicinity of Kragujevac through comprehensive soil sampling and analysis. Employing gamma spectrometry with high-purity germanium detectors (HPGe), we aim to discern the distribution of key radionuclides, including those from uranium and thorium series, as well as potassium-40. Gamma spectrometry with HPGe detectors was selected for its exceptional energy resolution and efficiency in detecting and quantifying radionuclides, making it a reliable choice for environmental monitoring. Additionally, the investigation extends to plant samples and commercially available and domestic milk, offering insights into radionuclide transfer mechanisms. Kragujevac is a significant location for this study due to its mixed urban-rural environment and proximity to industrial activities and agricultural practices, which may influence radionuclide distribution. Historical data suggest the potential for localized environmental contamination, necessitating ongoing monitoring to assess radiation levels and associated public health risks.

By elucidating the radioactivity profile of Kragujevac's surroundings, this research aims to provide valuable data for assessing population exposure to gamma radiation and internal radiation through food consumption. Such insights are essential for informing radiation protection measures and safeguarding public health in urban environments.

MATERIALS AND METHODS

From March to August 2024, a comprehensive sampling campaign was undertaken to assess the radioactivity levels in various environmental matrices within the study area. A total of ten soil samples were meticulously collected, representing both rural and urban environments in equal measure. Surface layers of soil, including vegetation and other debris, were carefully removed to minimize contamination and ensure that samples were obtained from the underlying soil matrix (IAEA, 2004). A map delineating the researched area and the sampling localities is provided in Figure 1. This balanced approach aimed to capture potential differences in radioactivity profiles between these distinct settings.

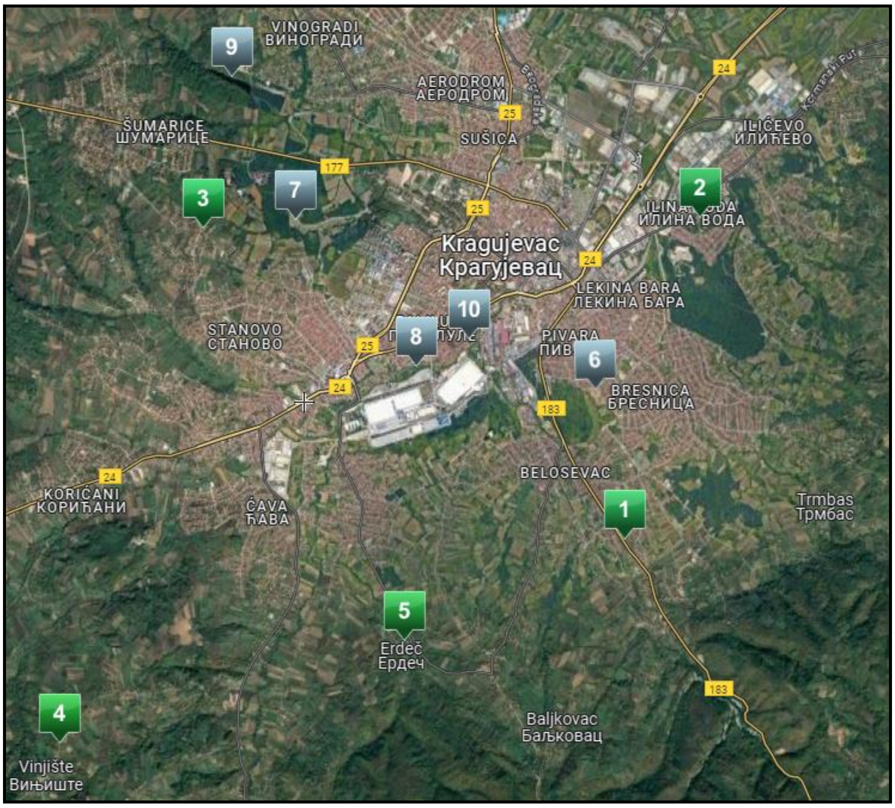

Figure 1 The map of sampling locations in the city of Kragujevac and surrounding villages.
a) green markers indicate rural areas; b) gray markers represent urban areas

Furthermore, the sampling strategy included eight plant samples, with five sourced from rural locations and cultivated using organic practices (strawberry, cherry, plum, tomato, and cucumber), and three sourced from urban locations using inorganic cultivation methods (spinach, pepper, and zucchini). This consideration allowed for an exploration of how different agricultural methods might influence the uptake of radionuclides by plants, thereby affecting food chain dynamics and potential human exposure pathways. Table 1. provides a detailed overview of the distribution of plant samples, categorized by their location (rural or urban) and the type of cultivation method used (organic or inorganic).

Table 1. Plant sample distribution by location and cultivation type.

Location	***Soil Sample***	***Plant Type***
1	Rural	Strawberry (Organic)
2	Rural	Cherry (Organic)
3	Rural	Plum (Organic)
4	Rural	Tomato (Organic)
5	Rural	Cucumber (Organic)
6	Urban	Spinach (Inorganic)
7	Urban	Pepper (Inorganic)
8	Urban	Zucchini (Inorganic)

In addition to soil and plant samples, four milk samples were included in the study: two sourced domestically and two purchased commercially. The domestic milk was sampled from locations 1 and 5, the same sites from which the plant samples were collected, ensuring consistency in the source of the samples for radionuclide contamination assessment. Milk serves as an essential component of the human diet and can act as a conduit for the transfer of radionuclides from the environment to consumers (SHIKHA *et al*., 2024).

Gamma spectrometry analysis

The samples were prepared according to standard procedures for radionuclide determination (IAEA, 2004). They were placed in containers with Marinelli geometry, allowing for accurate measurement of specific activities. Gamma spectrometry, a widely employed technique for radioactivity analysis, was utilized to quantify the levels of key radionuclides, including potassium-40, uranium-238, thorium-232, and cesium-137. A high-purity germanium semiconductor detector (GEM30-70, ORTEC) with a relative efficiency of 30% was employed for precise measurements.

Each sample underwent gamma spectrometry analysis for a predetermined measurement time of 172800 seconds (approximately 48 hours). This extended measurement duration ensured sufficient statistical precision in determining specific activities of radionuclides present in the samples. The computer software MAESTRO 2 was used to perform spectrum analysis.

RESULTS AND DISCUSSION

The activity concentrations of key radionuclides (^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs) varied across different sample types and locations within the study area. The results of the specific activity of natural radionuclides and ^{137}Cs are presented in Table 2. The data collected provides valuable insights into the distribution and potential sources of these radionuclides in different settings, as well as their uptake by various plants and the possible implications for food safety.

Table 2. Mean activity concentrations [Bq/kg] for soil and milk samples with specific activity concentrations for each plant type.

Type of Sample ↓	**Act. conc. →**	***^{226}Ra (Bq/kg)***	***^{232}Th (Bq/kg)***	***^{40}K (Bq/kg)***	***^{137}Cs (Bq/kg)***
Soil (rural)		37.2 ± 9.2	56.2 ± 15.1	670.0 ± 75.0	52.7 ± 13.3
Soil (urban)		29.3 ± 3.6	44.1 ± 5.6	660.0 ± 92.0	17.9 ± 12.8
Strawberry (Organic)		4.8 ± 1.5	5.2 ± 1.8	100.0 ± 20.0	ND*
Cherry (Organic)		6.3 ± 2.7	7.1 ± 3.1	120.0 ± 30.0	3.5 ± 0.8
Plum (Organic)		5.1 ± 1.9	5.8 ± 2.2	110.0 ± 25.0	ND
Tomato (Organic)		5.6 ± 2.3	6.4 ± 2.9	130.0 ± 35.0	2.1 ± 0.5
Cucumber (Organic)		6.8 ± 2.4	7.5 ± 3.0	140.0 ± 40.0	ND
Spinach (Inorganic)		4.9 ± 1.7	5.3 ± 2.0	150.0 ± 45.0	1.8 ± 0.4
Pepper (Inorganic)		6.1 ± 2.8	6.9 ± 3.3	160.0 ± 50.0	ND
Zucchini (Inorganic)		5.3 ± 2.0	6.0 ± 2.5	155.0 ± 48.0	2.7 ± 0.6
Milk (purchased)		2.5 ± 1.1	3.0 ± 1.2	175.0 ± 50.0	9.5 ± 2.8
Milk (domestic)		ND	ND	67.0 ± 10.0	ND

*ND – not detected

The soil samples, divided into rural and urban categories, exhibit noticeable differences in radionuclide concentrations. Rural soil samples show higher mean concentrations of ^{226}Ra (37.2 Bq/kg) and ^{232}Th (56.2 Bq/kg) compared to urban soil (29.3 Bq/kg for ^{226}Ra and 44.1

Bq/kg for ^{232}Th). This disparity may be attributed to the natural geological differences between rural and urban areas, where rural soils may have a higher natural radionuclide content due to underlying rock types or past agricultural practices (AMARAL *et al.*, 1992; GULAN *et al.*,2013; AJAYI and IBIKUNLE, 2013). Moreover, the significantly higher ^{137}Cs concentration in rural soil (52.7 Bq/kg) compared to urban soil (17.9 Bq/kg) could indicate past fallout events or specific local contamination sources that predominantly affected rural areas (KRSTIĆ *et al.*, 2004). Regarding potassium-40, both rural and urban soil samples exhibit comparable concentrations, with rural soil showing a mean activity of 670.0 Bq/kg and urban soil 660.0 Bq/kg. These similar levels reflect the ubiquitous nature of potassium-40 in the environment, as it is a naturally occurring radionuclide present in both types of soils due to the potassium content in minerals (UNSCEAR, 2000). In crops like cherries and tomatoes, detectable levels of ^{137}Cs (3.5 Bq/kg and 2.1 Bq/kg, respectively) raise questions about potential health risks. While these levels are relatively low, prolonged exposure through consumption could lead to cumulative internal radiation doses. To contextualize these findings, it is essential to compare them with permissible safety limits for ^{137}Cs in food products, which vary by region but are typically around 1,000 Bq/kg for most foods (WHO, 2021). The detected levels in this study fall significantly below these thresholds, indicating minimal immediate health risks. However, the presence of ^{137}Cs in these crops highlights the need for continued monitoring and risk assessment to ensure long-term food safety.

The plant samples, categorized by type (organic vs. inorganic), reveal varying levels of radionuclide uptake. Organic plant samples such as strawberries, cherries, plums, tomatoes, and cucumbers show generally low concentrations of ^{226}Ra and ^{232}Th, with values ranging between 4.8 Bq/kg in strawberries and 6.8 Bq/kg in cucumbers for ^{226}Ra, and from 5.2 Bq/kg in strawberries to 7.5 Bq/kg in cucumbers for ^{232}Th. The ^{40}K levels ranged from 100 Bq/kg in strawberries to 140 Bq/kg in cucumbers, reflecting its natural abundance in plants as a vital nutrient. However, the detection of ^{137}Cs in specific organic crops, such as cherries (3.5 Bq/kg) and tomatoes (2.1 Bq/kg), is particularly noteworthy. The presence of ^{137}Cs, even in low concentrations, could suggest ongoing environmental contamination or historical residues still present in the soil, leading to uptake by these plants (MOLLAH, 2014).

Inorganic crops like spinach and pepper also demonstrate similar trends, with ^{226}Ra and ^{232}Th concentrations within the expected range for plants. The detection of ^{137}Cs in spinach (1.8 Bq/kg) and zucchini (2.7 Bq/kg) highlights the potential variability in radionuclide uptake depending on the type of plant and its growing conditions (MOHAMMED and HAULE, 2018; ELYWA, 2019). Although zucchinis are categorized as inorganic in this study, their levels of ^{137}Cs are comparable to some organic plants, indicating that both types of crops can accumulate this radionuclide. The relatively low levels of ^{137}Cs in these samples are still significant, as they could contribute to the overall dietary intake of radionuclides, particularly in populations with high consumption of these vegetables.

The milk samples present an interesting contrast between purchased and domestic sources. The purchased milk shows detectable levels of ^{226}Ra (2.5 Bq/kg), ^{232}Th (3.0 Bq/kg), ^{40}K (175.0 Bq/kg), and ^{137}Cs (9.5 Bq/kg). The presence of ^{137}Cs in purchased milk is particularly concerning, as it indicates that the feed or water supply for commercially raised cattle may be contaminated. Potential differences in farming practices could explain these higher radionuclide levels in purchased milk. Commercial cattle often rely on mass-produced feed and water sources that may originate from regions with varying levels of contamination. On the other hand, domestic milk samples exhibit non-detectable levels of most radionuclides except for ^{40}K (66.9 Bq/kg), which is expected due to its natural presence in milk as part of normal biological processes (HOWARD *et al.*,2009; DESIMONI *et al.*,2009). The absence of detectable ^{137}Cs in domestic milk might suggest that the cattle in this scenario are raised in environments with lower contamination levels, possibly due to better-controlled feeding practices or cleaner environmental conditions. According to international guidelines, such as those from the FAO Codex Alimentarius (WHO, 2021), the maximum permissible concentration of ^{137}Cs in food products, including milk, ranges from 100 to 1000 Bq/kg, depending on the region. The fact that no detectable levels of ^{137}Cs were found in the milk samples collected further supports the conclusion that cesium contamination is well within acceptable limits, ensuring that the milk is safe for consumption.

The data indicates that environmental and agricultural practices significantly influence the distribution and uptake of radionuclides. The higher radionuclide concentrations in rural soil might be due to the natural geology or historical agricultural practices, which could have enhanced the radionuclide content in these areas. The presence of ^{137}Cs in both plant and milk samples raises concerns about ongoing environmental contamination and its potential impact on food safety. Although the detected levels are relatively low, they warrant continuous monitoring to ensure they remain within safe limits for human consumption. The differences between organic and inorganic plant samples suggest that while both cultivation practices result in similar uptake for certain radionuclides, the specific plant type and growing conditions play a crucial role in determining the levels of radionuclides present. The detected ^{137}Cs concentrations, even at low levels, highlight the importance of ongoing surveillance, particularly in regions with a history of radioactive contamination (KALKAN *et al*., 2021; PETROVIĆ *et al*., 2013 ; JANKOVIĆ *et al*., 2023). In summary, this table underscores the need for ongoing environmental monitoring and the potential impact of environmental contamination on food products. While the detected levels of radionuclides are generally low, their presence in both plant and milk samples indicates the need for careful management of agricultural practices and food safety protocols to minimize human exposure to these contaminants. To address ongoing contamination concerns, implementing targeted monitoring programs and promoting the use of clean feed and water sources for livestock, as well as controlled cultivation practices, could help mitigate radionuclide uptake in food products.

CONCLUSION

The study highlights the variability in radionuclide concentrations across soil, plant, and milk samples, with rural soils showing higher levels of ^{226}Ra and ^{137}Cs, likely due to natural geology and historical fallout. The presence of ^{137}Cs in certain crops, even at low levels, indicates ongoing environmental contamination that requires continuous monitoring. Differences in radionuclide uptake between organic and inorganic plant samples underscore the influence of agricultural practices. The detection of ^{137}Cs in purchased milk raises concerns about commercial food safety, while domestic milk showed minimal contamination. This research underscores the importance of continued monitoring and Future research could focus on exploring the long-term effects of radionuclide contamination on a wider range of crops and developing more efficient agricultural practices to minimize radionuclide uptake. targeted mitigation strategies to protect public health in areas with known contamination.

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References:

[1] AJAYI, O.S., IBIKUNLE, S.B. (2013): Radioactivity of Surface Soils from Oyo State, South Western Nigeria. *International Journal of Radiation Research* **11** (4): 271–278.

[2] AMARAL, E.C.S., ROCHEDO, E.R.R., PARETZKE, H.G., FRANCA, E.P. (1992): The Radiological Impact of Agricultural Activities in an Area of High Natural Radioactivity. *Radiation Protection Dosimetry* **45** (1–4): 289–292 pp. doi: 10.1093/rpd/45.1-4.289

[3] CAMPBELL, J.E., MURTHY, G.K., STRAUB, C.P., LEWIS, K.H., TERRILL, J.G. (1961): Radionuclides in Milk. *Journal of Agricultural and Food Chemistry* **9** (2): 117–122. doi: 10.1021/jf60114a008

[4] DESIMONI, J., SIVES, F., ERRICO, G., MASTRANTONIO, J. (2009): Activity levels of gamma-emitters in Argentinean cow milk. *Food Composition and Analysis* **22** (3–4): 250–253. doi: 10.1016/j.jfca.2008.10.024

[5] DJELIC, G., KRSTIC, D., STAJIC, J.M., MILENKOVIC, B., TOPUZOVIC, M., NIKEZIC, D., VUCIC, D., ZEREMSKI, T., STANKOVIC, M., KOSTIC, D. (2016): Transfer factors of natural radionuclides and 137Cs from soil to plants used in traditional medicine in central Serbia. *Journal of Environmental Radioactivity* **158-159**: 81–88. doi: 10.1016/j.jenvrad. 2016.03.028

[6] ELYWA, M. (2019): Determination of Radioactivity Concentration and Assessment of Absorbed Effective Dose of Edible Plant and Soil Samples. *Arab Journal of Nuclear Sciences and Applications,* **52** (3): 56–63. doi: 10.21608/ajnsa.2019.5754.1132

[7] WHO - World Health Organization & Food And Agriculture Organization Of The United Nations (2021): Discussion paper on the radioactivity in food and feed (including drinking water) in non-emergency situations. *Codex Alimentarius Commission*, 14th Session (CX/CF 21/14/14).

[8] GIRI, S., SINGH, G., JHA, V.N., TRIPATHI, R.M. (2011): Risk assessment due to ingestion of natural radionuclides and heavy metals in the milk samples: a case study from a proposed uranium mining area, Jharkhand. *Environmental Monitoring and Assessment* **175**: 157–166. doi: 10.1007/s10661-010-1502-8

[9] GULAN, L.R., BOCHICCHIO, F., CARPENTIERI, C., MILIĆ, G.A., STAJIĆ, J.M., KRSTIĆ, D.Ž., STOJANOVSKA, Z.A., NIKEZIĆ, D.R., ŽUNIĆ, Z.S. (2013): High Annual Radon Concentration in Dwellings and Natural Radioactivity Content in Nearby Soil in Some Rural Areas of Kosovo and Metohija. *Nuclear Technology and Radiation Protection* **28** (1): 60–67.

[10] HOWARD, B.J., BERESFORD, N.A., BARNETT, C.L., FESENKO, S. (2009): Quantifying the Transfer of Radionuclides to Food Products from Domestic Farm Animals. *Journal of Environmental Radioactivity* **100** (9): 767–773. doi: 10.1016/j.jenvrad.2009.03.010

[11] IAEA (2004): Soil sampling for environmental contaminants. IAEA-TECDOC-1415. https://www.iaea.org/search/google/Publication%202004

[12] JANKOVIĆ, M., JELIĆ, I., RAJAČIĆ, M., KRNETA N.J., VUKANAC, I., DIMOVIĆ, S., SARAP, N., ŠLJIVIĆ-IVANOVIĆ, M. (2023): Distribution of Natural Radionuclides and ^{137}Cs in Urban Soil Samples from the City of Novi Sad, Serbia—Radiological Risk Assessment. *Toxics,* **11** (4): 345. doi: 10.3390/toxics11040345

[13] KALKAN, S.K., FORKAPIĆ, S., MARKOVIĆ, B.S., GAVRILOV, B.M., BIKIT-SCHROEDER, K., MRĐA, D., RADAKOVIĆ, G.M., TOŠIĆ, R. (2021): Deposition of ^{137}Cs and Precipitation

Distribution in Vojvodina, Northern Serbia After the Chernobyl Accident. *Chemosphere* **264** (2): 128471. doi: 10.1016/j.chemosphere.2020.128471

[14] KEUM, D.K., LEE, H., KANG, S.H., JUN, I., CHOI, Y.H., LEE, C.W. (2007): Predicting the transfer of 137137Cs to rice plants by a dynamic compartment model with a consideration of the soil properties. *Journal of Environmental Radioactivity* **92**(1): 1–15. doi: 10.1016/j.jenvrad.2006.08.007

[15] KRSTIĆ, D., NIKEZIĆ, D., STEVANOVIĆ, N., JELIĆ, M. (2004): Vertical profile of 137Cs in soil. *Applied Radiation and Isotopes* **61** (6): 1487–1492. doi: 10.1016/j.apradiso.2004.03.11

[16] MOHAMMED, N., HAULE, B. (2018): Assessment of Heavy Metal and Radioactivity Levels in Spinach (Spinacia oleracea) Grown at Bahi Wetlands in Dodoma Region. *Tanzania Journal of Science* **44** (1): 169–178.

[17] MOLLAH, A.S. (2014): Radionuclide Uptake from Soil to Plants: Influence of Soil Classification. In: Gupta, D., Walther, C. (eds) *Radionuclide Contamination and Remediation Through Plants*. Springer, Cham. doi: 10.1007/978-3-319-07665-2_3

[18] MONGED, M.H.E., HASSAN, H.B., EL-SAYED, S.A. (2020): Spatial Distribution and Ecological Risk Assessment of Natural Radionuclides and Trace Elements in Agricultural Soil of Northeastern Nile Valley, Egypt. *Water, Air, and Soil Pollution* **231** (7): 231–338. doi: 10.1007/s11270-020-04678-9

[19] PETROVIĆ, J., ĆUJIĆ, M., ĐORĐEVIĆ, M., DRAGOVIĆ, R., GAJIĆ, B., MILJANIĆ, Š., DRAGOVIĆ, S. (2013): Spatial Distribution and Vertical Migration of ^{137}Cs in Soils of Belgrade (Serbia) 25 Years After the Chernobyl Accident. *Environmental Science: Processes & Impacts* **15** (6): 1279–1289. doi: 10.1039/C3EM00084B

[20] SHIKHA, P., KABIR, M.M., DEWAN, M.J., KHANDAKER, M.U., YEASMIN, S. (2024): Evaluation of radioactivity concentration in farm fresh milk and concomitant dose to the consumer. *Heliyon* **10** (9): e30454. doi: 10.1016/j.heliyon.2024.e30454

[21] STÄGER, F., ZOK, D., SCHILLER, A.K., FENG, B., STEINHAUSER, G. (2023): Disproportionately High Contributions of 60 Year Old Weapons-137Cs Explain the Persistence of Radioactive Contamination in Bavarian Wild Boars. *Environmental Science and Technology* **57** (36): 13601–13611. doi: 10.1021/acs.est.3c03565

[22] UNSCEAR (2008): United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly. United Nations, New York

[23] UNSCEAR (2000): Report to the general assembly. Sources and efects of ionizing radiation, vol I, Annex B: exposures from natural radiation sources. United Nations, New York

[24] ZHU, Y.G., SHAW, G. (2000): Soil Contamination with Radionuclides and Potential Remediation. Chemosphere **41** (1–2): 121–128. doi: 10.1016/S0045-6535(99)00398-7