

The impact of natural radioactivity in animal products of ruminants on the annual effective dose of population

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Abstract

Activity concentration of ^{238}U , ^{226}Ra and ^{40}K were analyzed in samples of milk, meat, cow cheese and sheep cheese from the areas of three municipalities: Livno, Kakanj and Hadžići for the purpose of assessment of the annual effective dose by ingestion of animal products of ruminants.

Burned samples of milk and meat from cow and sheep were measured by modified gamma spectrometry method. High resolution HPGe detector with additional electronic equipment had been used for that purpose. All results were calculated on the fresh weight basis. Obtained results for samples of cow milk were in ranges: 0.010–0.020 Bq kg⁻¹ for ^{238}U ; 0.014–0.060 Bq kg⁻¹ for ^{226}Ra and 37.2–67.5 Bq kg⁻¹ for ^{40}K . Sheep milk showed the following ranges of obtained results: 0.007–0.033 Bq kg⁻¹ for ^{238}U ; 0.010–0.045 Bq kg⁻¹ for ^{226}Ra and 30.7–73.3 Bq kg⁻¹ for ^{40}K . Levels of observed radionuclides (^{238}U , ^{226}Ra and ^{40}K) in samples of cow cheese were moving between 0.010–0.070, 0.01–0.10, and 27.3–58.9 Bq kg⁻¹. Activity concentrations of ^{238}U , ^{226}Ra and ^{40}K in samples of cow meat were: 0.010–0.023 Bq kg⁻¹; 0.018–0.060 Bq kg⁻¹ and 66.2–96.9 Bq kg⁻¹, respectively. Levels of ^{238}U , ^{226}Ra and ^{40}K in samples of sheep meat were: 0.010–0.024 Bq kg⁻¹; 0.014–0.035 Bq kg⁻¹ and 65.2–99.5 Bq kg⁻¹, respectively. Calculated annual effective doses by ingestion of observed samples were in range 0.055–0.064 mSv.

Obtained results indicated acceptable levels of investigated natural radionuclides as well as correct radiation-hygienic status of animal products of ruminants from selected municipalities. Estimated annual effective doses for ingestion of animal products of ruminants were in the range of world average values.

Keywords

natural radionuclides – ruminants – effective dose

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Introduction

For the purpose of assessment of radiation risk of population by natural radiation, it is necessary to perform, among the other, the assessment of the annual effective dose of natural radionuclides by ingestion of agricultural and animal products.

Ingestion of natural radionuclides depends of the food consumption as well as of the radionuclides activity concentrations in specific food. World Health Organization - WHO (16) and Food and Agriculture Organization of the United Nations - FAO (2) reported the average annual consumption of the food products. Quantities of the particular food in annual diet significantly differ depending of the feeding practices that prevailing in specific region.

Activity concentration of natural radionuclides in different food categories are also very variable parameter who primary depends of background values of soil, potential impact of NORM industries and prevailing agricultural practices.

Many studies performed in areas of increased naturally background as well as in surroundings of industries who induce technologically enhanced natural radioactivity (TENORM – technologically enhanced naturally occurring radioactive materials) were pointed out on elevated levels

of natural radionuclides in agricultural and animal products that led to increasing of the annual effective doses by ingestion as well as to increased radiation risk of population living in investigated areas (1,3,4,5,9,11). Review of literature references points out on the conclusion that radiation risk by ingestion of the food and water is variable quantity and have to be considered in context of dominating factors that prevailing in particular area. Having regard that fact, UNSCEAR (12) reported average value of annual effective dose by ingestion of 0.3 mSv with range 0.2–0.8 mSv.

According to mentioned, assessment of impact of natural radioactivity on the annual effective dose of population by ingestion of animal products of ruminants was investigated at three areas with elevated levels of natural radioactivity.

Material and Methods

Study was performed in three areas: surroundings of the coal mine Tušnica-Drage, municipality Livno; surroundings of the coal-fired power plant Kakanj as well as surroundings of the industrial facility in municipality Hadžići, which was target of NATO strikes in 1995. Localities were chosen on the base of presence of industrial facilities who lead to

TENORM or on the base of doubt for presence of depleted uranium (DU).

Composite samples of meat, milk and cheese were taken from villages in surroundings of mentioned facilities. Study was focused on the animal products of ruminants (cows and sheep).

Preparing of samples for measuring was included their cleaning, weighing, homogenization, drying and burning in muffle furnace below 400 °C, as described in IAEA publication (6). The samples were kept for 120 days in order to establish the secular equilibrium between ^{238}U and ^{234}Th .

Gamma spectrometry measurements were performed on vertical coaxial HPGe detector, POP-TOP, p-type, produced by ORTEC, model GEM 30P4 with relative efficiency 30% and resolution 1.85 keV at energy 1.33 MeV.

Activity concentrations of the observed radionuclides (^{238}U , ^{232}Th , ^{226}Ra and ^{40}K) were calculated from their gamma lines as well as from gamma lines of their daughter products. Activity concentrations of ^{238}U were calculated from ^{234}Th energy at 63 keV as well as from the energy of $^{234\text{m}}\text{Pa}$ at 1001 keV. Levels of ^{232}Th were calculated from the energies of the daughter product ^{228}Ac (911 and 967 keV) as well as from the energy of daughter product ^{208}Tl (583 and 2614 keV). Activities of the ^{226}Ra were obtained from energies of ^{214}Pb (295 and 352 keV) and energies of ^{214}Bi (609 and 1764 keV). Levels of ^{40}K were calculated from its gamma energy at 1461 keV. Measuring time was 80 000 seconds.

For measuring activities of observed radionuclides that were below the detection limits, method of comparative measuring with standard sample (IAEA 414) with low levels of natural radionuclides was used. For that purpose it was necessary to provide the same measuring parameters related to mass, density, geometry and time of measuring. Activity concentration was calculated according to expression:

$$Au = \frac{Nu}{Ns} As$$

Where:

- Au – activity concentration of particular radionuclide at appropriate energy in the observed sample
- Nu – number of counts with subtracted background counts at appropriate energy in the observed sample
- Ns – number of counts with subtracted background counts at appropriate energy in the standard sample
- As – activity concentration of particular radionuclide at appropriate energy in the standard sample.

Results were calculated on the fresh weight basis (Bq kg^{-1} FW).

Annual effective doses were calculated according the expression:

$$Ex = Cx m DCFx$$

Where are:

- Ex – annual effective dose (mSv year^{-1})
- Cx – activity concentration of the particular radionuclide (Bq kg^{-1})

- m – yearly consumption of the particular food products (cow milk 70 L, sheep milk 3 L, cow cheese 10 kg, sheep cheese 3 kg, beef 50 kg, sheep meat 15 kg)
- DCFx – dose conversion factor for particular radionuclide – nSv / Bq (45 for ^{238}U , 280 for ^{226}Ra and 6.2 for ^{40}K)

Results

Average activities of ^{238}U and ^{226}Ra in animal products of ruminants are presented in Table 1. Results for ^{232}Th are not shown because of their low values. Average annual effective doses by ingestion for population at observed localities are shown in Tables 2-4.

Discussion and conclusion

Levels of ^{238}U and ^{226}Ra obtained in the study showed good agreement with the results recorded in Germany but slightly higher than those recorded in other European countries. Activities of ^{232}Th in all observed samples were below 0.01 Bq kg^{-1} what was in accordance with the results from other countries. Levels of ^{40}K were in the range of average values reported for European countries (12).

Statistical analysis of results showed low standard deviation for ^{238}U and ^{226}Ra (0.0006–0.07) and higher values for ^{40}K (3.3–21.4), which was indicated on the differences in nutritional requirements for potassium between different species of ruminants, as well as variability of ^{40}K distribution into different kinds of animal products.

Average activity concentration of ^{238}U and ^{226}Ra in cow milk samples from observed localities were presented in Table 1. The highest average activity concentrations of ^{238}U and ^{226}Ra in cow milk were recorded in Livno and the lowest ones in Kakanj. Generally, the higher levels of ^{226}Ra in animal products compared with uranium are caused by preferential intake of radium and its possible analogy with calcium which were reported by several authors (8,13,14,15). Levels of ^{40}K were under the world average values for observed kinds of samples (12).

Elevated levels of ^{238}U and ^{226}Ra in animal products of ruminants recorded at locality Livno were caused by increased levels of mentioned radionuclides in samples of soil and roughage feed which was reported by Saracevic et al. (10). Activities of ^{40}K in cow milk from Livno were the highest in the study, despite the fact that its concentration in soil and roughage feed were not high.

Average levels of observed radionuclides in animal products from Kakanj showed good agreement with the results of Samek et al. (7). Impact of the ash released from facility was significant for the food chain of ruminants. Increased levels of ^{238}U and ^{226}Ra in animal products from Kakanj were caused by the deposition of the ash from facility on the surface of roughage feed (grass and hay) who were the main component of ruminants diet, what was reported by Gradasevic (5).

Experimental work at locality Hadžići was focused on the determining of increased levels of ^{238}U what would be evidence of presence of depleted uranium in animal products of ruminants. Obtained results showed low levels

Table 1. Average activity concentrations of ^{238}U , ^{226}Ra and ^{40}K in animal products of ruminants (Bq L^{-1} , Bq kg^{-1} fresh weight)

	Livno			Kakanj			Hadžići		
	^{238}U	^{226}Ra	^{40}K	^{238}U	^{226}Ra	^{40}K	^{238}U	^{226}Ra	^{40}K
Cow milk	0.016	0.050	53.0	0.011	0.034	48.3	0.012	0.036	44.7
Sheep milk	0.018	0.032	46.4	0.011	0.017	51.0	0.010	0.013	68.6
Cow cheese	0.046	0.080	47.5	0.017	0.053	39.6	0.017	0.053	31.1
Cow meat	0.020	0.039	83.9	0.014	0.027	82.3	0.012	0.028	72.2
Sheep meat	0.017	0.028	86.4	0.012	0.015	73.7	0.010	0.015	87.5

Table 2. Annual effective dose by ingestion of animal products of ruminants for population in area of town Livno (nSv)

Animal product	Annual effective dose nSv			Total annual effective dose nSv
	^{238}U	^{226}Ra	^{40}K	
Cow milk	50	960	22959	23969
Sheep milk	2	27	863	892
Cow cheese	21	224	2945	3190
Sheep cheese	9	168	580	757
Beef	45	546	26009	26600
Sheep meat	12	118	8035	8165
Total	139	2043	61391	63573

Table 3. Annual effective dose by ingestion of animal products of ruminants for population in area of town Kakanj (nSv)

Animal product	Annual effective dose nSv			Total annual effective dose nSv
	^{238}U	^{226}Ra	^{40}K	
Cow milk	35	666	20962	21663
Sheep milk	2	14	949	965
Cow cheese	8	140	2455	2603
Sheep cheese				
Beef	32	378	25513	25923
Sheep meat	8	63	6798	6869
Total	85	1261	56677	58023

Table 4. Annual effective dose by ingestion of animal products of ruminants for population in area of town Hadžići (nSv)

Animal product	Annual effective dose nSv			Total annual effective dose nSv
	^{238}U	^{226}Ra	^{40}K	
Cow milk	38	706	19417	20161
Sheep milk	1	11	1277	1289
Cow cheese	8	148	1927	2083
Sheep cheese				
Beef	27	392	22373	22792
Sheep meat	7	63	8136	8206
Total	81	1320	53130	54531

of ^{238}U with high uncertainty (40%) which was not allowed the assessment of the DU presence in observed samples.

The highest annual effective dose by ingestion of animal products was recorded at locality Livno with calculated value of 0.064 mSv. The highest contribution was coming from ^{40}K (96.5%) whilst uranium series of radionuclides, ^{238}U and ^{232}Th , contributed with 3.2% and 0.3%, respectively.

According to UNSCEAR report (12), animal products of ruminants contribute to the total annual diet of population with 155 kg (15%). Under the hypothesis that annual effective dose by ingestion of animal products of ruminants in Livno (0.064 mSv) represents 15% of the total annual ingestion dose, calculation on the total annual dose will give the value of 0.426 mSv for locality Livno. Calculated value gave the good agreement with result of Saracevic et al. (10) who obtained the annual effective dose by ingestion of 0.412 mSv for the same locality. Calculated annual effective dose by ingestion was slightly higher than average value (0.3 mSv) but under the range (0.2–0.8 mSv) given in UNSCEAR report (12).

Calculated annual effective dose by ingestion of animal products of ruminants for population in area of town Kakanj was 0.058 mSv. The highest contribution was also came from ^{40}K (97.7%) while contribution of the ^{226}Ra (2.15%) and ^{238}U (0.15%) were significantly lower than those recorded in area of town Livno. Total annual effective dose by ingestion estimated by the method described for Livno, was 0.386 mSv. Obtained value was higher than those reported by Samek (7) probably because of the fact that sampling was performed much closer to coal-fired power plant Kakanj than in the previous work. Calculated value of annual effective dose was slightly higher than average value reported by UNSCEAR (12) probably because of the higher impact of the deposited ash from facility on the surface of the grass (hay) who were the main component of the diet of ruminants. The calculated annual effective dose was still under the range of annual effective doses by ingestion reported by UNSCEAR (12).

Annual effective dose by ingestion of animal products of ruminants for population in the area Hadžići was 0.055 mSv. The highest contribution was coming from ^{40}K (97.4%) with significantly lower values recorded for ^{226}Ra (2.4%) and ^{238}U (0.15%). Recorded values was not pointed out on the significant impact of uranium on the annual effective dose by ingestion for population in Hadžići, what was evidence that eventually presence of the depleted uranium in the area of town Hadžići was not caused the contamination of animal products of ruminants. Calculated annual

effective dose by ingestion based on the contribution of the animal products of ruminants (15% of total diet) was 0.364 mSv what was slightly higher than UNSCEAR reported average value of 3 mSv, but still in the range of reported values (12).

The highest value of annual effective dose by ingestion of the animal products of ruminants was recorded at locality Livno (0.064 mSv), while the similar values were obtained in Kakanj (0.058) and Hadžići (0.055 mSv). The main contribution to the dose was generally coming from ^{40}K (97.2%) with significantly lower contribution of ^{226}Ra (2.6%) and ^{238}U (0.16%).

Activity concentrations of natural radionuclides ^{238}U , ^{226}Ra and ^{40}K recorded in samples of animal products of ruminants at investigated localities were not showed significant enhancement compared with world average values. Values of estimated annual effective dose by ingestion were also under the range of the world average values. Obtained results were indicated on the acceptable radiation-hygiene validity of animal products of ruminants from observed areas.

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Utjecaj prirodne radioaktivnosti u animalnim proizvodima preživara na godišnju efektivnu dozu populacije

Sažetak

Uvod i ciljevi

Procjena radijacionog opterećenja populacije nekog područja podrazumijeva, između ostalog, i procjenu opterećenja te populacije ingestijom poljoprivrednih i animalnih proizvoda toga područja. Ingestija prirodnih radionuklida u zavisnosti je od godišnje konzumacije hrane, kao i koncentracije aktivnosti prirodnih radionuklida u pojedinim namirnicama. Utvrđeno je da vrijednosti konzumacije pojedinih namirnica značajno variraju, u zavisnosti od prehrambenih navika koje prevladavaju u određenom regionu. Koncentracije aktivnosti prirodnih radionuklida u različitim kategorijama namirnica su također varijabilan parametar koji primarno zavisi od inicijalnih vrijednosti radionuklida u tlu, potencijalnom utjecaju industrija koje dovode do tehnološki izmijenjene prirodne radioaktivnosti i prevladavajućoj poljoprivrednoj praksi na nekom području.

Brojna istraživanja izvedena u područjima povišene prirodne radioaktivnosti, kao i područjima u okruženju industrija koje dovode do tehnološki povišene prirodne radioaktivnosti, ukazala su na povišene nivoe prirodnih radionuklida u poljoprivrednim i animalnim proizvodima i, posljedično, povećanje godišnjih ekvivalentnih doza ingestijom.

Pregledom literaturnih podataka, može se zaključiti da je radijacioni rizik ingestijom namirnica i vode varijabilna vrijednost, koja mora biti posmatrana u kontekstu prevladavajućih faktora na nekom području. U skladu sa tim, Naučni komitet Ujedinjenih nacija o efektima nuklearnog zračenja (UNSCEAR) objavio je prosječne vrijednosti godišnje efektivne doze ingestijom (0.3 mSv), s rasponom prihvatljivih vrijednosti (0.2–0.8 mSv).

U skladu s navedenim, procjena utjecaja prirodne radioaktivnosti na godišnju efektivnu dozu populacije ingestijom animalnih proizvoda preživara istraživana je na tri područja Bosne i Hercegovine s prethodno zabilježenim povišenim nivoima prirodnih radionuklida u tlu.

Materijal i metode

Istraživanje je izvedeno na tri područja: okolini rudnika Tušnica-Drage u Općini Livno; okolini termoelektrane Kakanj i okruženju bivšeg Tehničko-remontnog zavoda u Općini Hadžići, koji je bio meta NATO udara tokom 1995. godine. Lokaliteti su odabrani na osnovu prisustva industrija koje dovode do tehnološki izmijenjene prirodne radioaktivnosti ili na osnovu postojanja sumnje u prisustvo osiromašenog uranija.

Zbirni uzorci animalnih proizvoda preživara (mesa, mlijeka i sira krava i ovaca) uzimani su iz sela u okruženju navedenih postrojenja. Priprema i mjerenje uzoraka vršeni su prema standardnim procedurama. Rezultati su obračunavani

po 1 kg svježje mase uzorka.

Za procjenu godišnje efektivne doze ingestijom korišteni su podaci o godišnjoj konzumaciji animalnih proizvoda preživara, dobiveni anketom na istraživanim područjima. Godišnja efektivna doza dobijena je množenjem prosječnih nivoa radionuklida u ispitivanim uzorcima s godišnjom konzumacijom pojedinih animalnih proizvoda preživara i konverzionim faktorima za provođenje aktivnosti u efektivnu dozu, navedenim u izvještaju UNSCEAR (2000).

Rezultati i interpretacija

Rezultati dobijeni za uzorke kravljeg mlijeka bili su u rasponima: 0,010–0,020 Bq kg⁻¹ za ²³⁸U; 0,014–0,060 Bq kg⁻¹ za ²²⁶Ra i 37,2–67,5 Bq kg⁻¹ za ⁴⁰K. U ovčijem mlijeku zabilježeni su sljedeći nivoi ispitivanih radionuklida: 0,007–0,033 Bq kg⁻¹ za ²³⁸U; 0,010–0,045 Bq kg⁻¹ za ²²⁶Ra i 30,7–73,3 Bq kg⁻¹ za ⁴⁰K. Nivoi ²³⁸U, ²²⁶Ra i ⁴⁰K u kravljem siru bili su u rasponima: 0,010–0,070, 0,01–0,10, i 27,3–58,9 Bq kg⁻¹. Rezultati dobijeni za kravlje meso kretni su se u rasponima: 0,010–0,023 Bq kg⁻¹ (²³⁸U); 0,018–0,060 Bq kg⁻¹ (²²⁶Ra) i 66,2–96,9 Bq kg⁻¹ (⁴⁰K). Ovčije meso pokazalo je sljedeće raspone ispitivanih radionuklida (²³⁸U, ²²⁶Ra i ⁴⁰K): 0,010–0,024 Bq kg⁻¹; 0,014–0,035 Bq kg⁻¹ i 65,2–99,5 Bq kg⁻¹.

Najviše vrijednosti godišnje efektivne doze ingestijom animalnih proizvoda preživara zabilježene su na području Livna (0,064 mSv), dok su približno jednake vrijednosti dobijene na području Kakanja (0,058 mSv) i Hadžića (0,055 mSv). Vrijednosti godišnjih efektivnih doza ingestijom animalnih proizvoda preživara na istraživanim područjima bile su nešto više od prosječne vrijednosti navedene u izvještaju UNSCEAR-a (2000) od 3 mSv, ali u okviru raspona prihvatljivih vrijednosti (0,2–0,8 mSv) navedenih u istom izvještaju. Razlog viših vrijednosti doza na području Livna je prirodno povišen sadržaj ispitivanih radionuklida u tlu ovog područja. Na području Kakanja i Hadžića, dobijene više vrijednosti uzrokovane su blagim utjecajem pepela termoelektrane u Kakanju, odnosno viših vrijednosti ⁴⁰K u tlu na području Hadžića. Nije zabilježen utjecaj eventualno prisutnog osiromašenog uranija u tlu na području Hadžića na nivoe uranija u animalnim proizvodima preživara.

Zaključak

Koncentracije aktivnosti prirodnih radionuklida ²³⁸U, ²²⁶Ra i ⁴⁰K, zabilježene u uzorcima animalnih proizvoda preživara s istraživanih područja nisu pokazale povišene nivoe u poređenju s prosječnim svjetskim vrijednostima. Vrijednosti procijenjenih godišnjih efektivnih doza ingestijom bile su također u okviru prosječnih svjetskih vrijednosti. Dobijeni rezultati ukazali su na prihvatljivu radijaciono-higijensku ispravnost animalnih proizvoda preživara s istraživanih područja.