Activity concentrations of natural radionuclides in hay and grass from certain areas of Bosnia and Herzegovina

Nedžad Gradaščević1*, Lejla Saračević1, Davorin Samek1

Abstract
The amounts of natural radionuclides in the hay and grass from the city areas of Livno, Kakanj and Hadžići, and the mountain Vlašić, were investigated in the research paper. Activity concentration of natural radionuclides $^{238}$U, $^{232}$Th, $^{226}$Ra and $^{40}$K were calculated by gamma spectrometry with high resolution HPGe detector and additional equipment. Amounts of natural radionuclides (Bq kg$^{-1}$ fresh weight) in the hay samples were ranging from 0.1–2.6 for $^{238}$U, 0.01–0.6 for $^{232}$Th, 0.2–3.8 for $^{226}$Ra, and 275–817 for $^{40}$K. The following ranges of activity levels (Bq kg$^{-1}$ fresh weight) of $^{238}$U, $^{232}$Th, $^{226}$Ra and $^{40}$K were recorded in the grass samples: 0.06–0.70, 0.01–0.1, 0.11–1.1, 101–170, respectively. Obtained results did not point out at significantly elevated amounts of natural radionuclides in the samples of roughage feed. Recorded activity concentration of natural radionuclides in the samples of roughage feed indicated the acceptable amounts of natural radioactivity in the diet of ruminants in the observed areas.

Keywords
Natural radioactivity — roughage feed — ruminants

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Introduction
Areas with increased activity concentrations of natural radionuclides of U and Th series in the soil are also known as areas with increased amounts of natural radionuclides in the food chain. Besides, many industries like mining, phosphates processing, coal mining as well as coal power planting lead to accumulating big amounts of natural radionuclides ($^{226}$Ra, $^{238}$U, $^{210}$Pb, $^{232}$Th) or heavy metals (As, Pb, Cu, Zn) in the surface soil layers and surrounding environment. Especially important issue is the contamination of agricultural lands that provide the food, and thus contribute to radiation dose of population by ingestion (5). Contamination of roughage feed is commonly the main cause of contamination of ruminants and their products. Contamination of the plants is appearing by radionuclide deposition on the plant surface, or radionuclides have been absorbed by the root uptake. Resuspension of radionuclides from the soil surface is appearing due to impacts of rain, wind or grazing, which is of particular interest for the green pieces of plants (2). Significant variation in contamination level could be expected depending on: the plant species, parts of the plants, growth condition and plant processing before consuming (10). Removing of radionuclides from the plant surface is carried out by natural processes like wind dispersion or washing out by rainfall (3). The three main processes: interception, retention and translocation are connected in managing of contamination caused by the deposition on the plant surface (13). Plant contamination by the root uptake is more complex process prevailing in the later contamination period. Radiological assessment of food chain contamination requires also the knowledge of migration processes as well as the knowledge about radionuclides bioavailability. Bioavailability implies the availability of particular radionuclide for the root uptake. Numerous studies investigated the impact of physico-chemical soil properties on the radionuclide bioavailability (11,14,18,19,20). With regard to the high diversity of physico-chemical soil properties in Bosnia and Herzegovina, expected differences in bioavailability of radionuclides should lead to different amounts of natural radionuclides in the grass and hay as dominant feedstuffs in the diet of ruminants. According to the previously mentioned, the study was performed to investigate the amounts of natural radionuclides in roughage feed of ruminants from the selected areas of Bosnia and Herzegovina. The goal of the study was to assess impact of increased amounts of natural radionuclides in soil on potential radioactive contamination of dominant feedstuffs of ruminants.

Material and Methods
Hay and grass were sampled from the grasslands or supplied by the farmers in the town areas of Livno, Kakanj and Hadžići as well as the mountain Vlašić. Localities are selected according to recorded increased amounts of natural radionuclides produced by industrial facilities (Livno – coal mine, Kakanj - coal power plant). Town area of Hadžići
was selected in order to check for the possible presence of depleted uranium, whilst the mountain Vlačić area was the control locality without the external impact on natural radioactivity.

Ashed samples of the grass and hay were prepared according to the IAEA publication procedures (7).

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

Activity concentrations of the investigated radionuclides ($^{238}$U, $^{226}$Th, $^{226}$Ra and $^{40}$K) were calculated from their gamma lines as well as the gamma lines of their daughter products. Activity concentrations of $^{238}$U were calculated from $^{234}$Pa at 1001 keV as well as from the energy of $^{234}$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 daughter product $^{208}$Tl (583 and 2614 keV). Activities of the $^{226}$Ra were obtained from the 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 80000 seconds.

For measuring activities of observed radionuclides that were below the detection limits, a method of comparative measuring with standard samples (IAEA 414) with the 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 measurement. Activity concentration was calculated according to the equation:

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

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

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

### Results and discussion

Amounts of natural radionuclides ($^{238}$U, $^{232}$Th, $^{226}$Ra and $^{40}$K) in the samples of roughage feed are presented in Tables 1–4. Activity concentrations of $^{238}$U, $^{232}$Th, $^{226}$Ra and $^{40}$K in the samples of roughage feed from Livno are presented in Table 1. Increased levels of $^{238}$U and $^{226}$Ra in the samples of hay and grass in comparison with the control locality samples were caused by increased amounts of observed radionuclides in the soil as reported in the previous work (17). Considering that the sampling was performed near and far away from the coal mine, obtained results showed relatively low standard deviation (Table 1).

<table>
<thead>
<tr>
<th>Activity concentrations (Bq kg$^{-1}$) of $^{238}$U, $^{232}$Th, $^{226}$Ra and $^{40}$K in samples of hay and grass (fresh weight) from the town area of Livno</th>
</tr>
</thead>
<tbody>
<tr>
<td>Activity</td>
</tr>
<tr>
<td>-----------</td>
</tr>
<tr>
<td>$^{238}$U</td>
</tr>
<tr>
<td>Mean 0.2 0.3 0.4 4.87</td>
</tr>
<tr>
<td>Min 0.2 0.01 0.2 101</td>
</tr>
<tr>
<td>Max 0.8 0.06 1.1 441</td>
</tr>
</tbody>
</table>

Measured values for hay and grass showed normal hay/grass activity ratio in the range 3–4, which was equal to humid content in the observed samples. That finding indicated equilibrated activities in the hay and grass samples. Statistically significant positive correlation between $^{238}$U and $^{226}$Ra levels was found for hay samples ($R = 0.805$) as well as for grass samples ($R = 0.966$), which pointed out at the natural origin and normal distribution of these radionuclides. Obtained results showed that coal mine activities had no impact on uranium and radium levels in the samples of roughage, in particular with regard to similar amounts of the same radionuclides recorded in the vicinity and on long distance from the coal mine. Recorded low levels of thorium in the observed samples were expected in accordance with its low bioavailability, as reported in several studies (9,10,11,22,23).

Activity concentration of natural radionuclides in the samples of roughage feed from the vicinity of the coal power plant Kakanj, and statistical parameters are shown
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Table 3. Activity concentrations (Bq kg$^{-1}$) of $^{238}$U, $^{226}$Th, $^{226}$Ra and $^{40}$K in samples of hay and grass (fresh weight) from the town area of Hadžići

<table>
<thead>
<tr>
<th></th>
<th>$^{238}$U</th>
<th>$^{232}$Th</th>
<th>$^{226}$Ra</th>
<th>$^{40}$K</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hay</td>
<td>0.2 ± 0.1</td>
<td>0.2 ± 0.1</td>
<td>0.2 ± 0.1</td>
<td>385 ± 50.6</td>
</tr>
<tr>
<td>Grass</td>
<td>0.2 ± 0.1</td>
<td>0.2 ± 0.1</td>
<td>0.2 ± 0.1</td>
<td>135 ± 21.3</td>
</tr>
<tr>
<td>Mean</td>
<td>0.2</td>
<td>0.2</td>
<td>0.2</td>
<td>202.6</td>
</tr>
<tr>
<td>Min</td>
<td>0.1</td>
<td>0.1</td>
<td>0.1</td>
<td>153.8</td>
</tr>
<tr>
<td>Max</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td>275.3</td>
</tr>
<tr>
<td>SD</td>
<td>0.07</td>
<td>0.07</td>
<td>0.07</td>
<td>20.8</td>
</tr>
</tbody>
</table>

in Table 2. Obtained values varied depending on the wind direction and distance from the facility. Higher values were recorded at sites in close vicinity of the facility as well as at 5 km distance from the facility in down-wind direction. The main cause of increased amounts recorded in the hay and grass samples was deposition of ash originated from the facility, as reported in previous studies (15) as well as the resuspension of ash from the ash dump on the surface of the plants in surroundings.

Presence of the ash at the plant surface was confirmed by higher ash content in the burned samples, compared with the values from other localities. Having in mind that washing out of the samples was not performed. presence of the ash at the plant surface contributed to total recorded activity of hay and grass in the impact zone of the facility. The higher percentage of the ash was recorded in the samples of grass compared to hay, probably because of the higher humid content in grass, which increased ash adsorption at the grass surface. The second reason was dropping out of the ash during the hay drying. Statistical analysis of the results showed significantly positive correlation between activities of $^{238}$U and $^{226}$Ra in samples of hay and grass (R=0.96) compared with that of grass (R=0.56). This finding led to the conclusion that higher percentage of the ash from the facility caused disbalance of equilibrium between the aforementioned radionuclides in the samples of grass as well as generally higher levels of observed radionuclides calculated on the dry weight basis. Obtained results indicated that depositing ash from the coal power plant Kakanj influenced the food chain of ruminants.

Average activity concentrations of $^{238}$U, $^{226}$Th, $^{226}$Ra and $^{40}$K in the samples of roughage feed in the municipality Hadžići are presented in Table 3. Sampling of material was performed in surroundings of the facility targeted during the NATO strikes in 1995.

Results of measurements did not show significant increase of $^{238}$U in the hay samples compared with that from the control locality Vitovlje on the mountain Vlašić (Table 4). Activity concentrations of $^{232}$Th and $^{226}$Ra were also within the acceptable limits. Activity concentrations of $^{40}$K showed significant variability depending on the sampling place, and were in the range from 248-817 Bq kg$^{-1}$. Calculated on the dry weight basis. Statistical analysis of the results showed very low standard deviation for $^{238}$U, $^{232}$Th, $^{226}$Ra and $^{40}$K, which were in the range from 0.06 -0.37 compared to $^{40}$K, which demonstrated extremely high standard deviation of 206.8. Correlation analysis between activities of $^{238}$U and $^{226}$Ra in the samples of hay demonstrated low correlation coefficient (R=0.56), which pointed out at the possibility of presence of the low concentrations of depleted uranium. In the contrary, high correlation coefficient in the samples of grass (R=0.85) indicated the regular ratio and origin of the mentioned radionuclides.

It is necessary to point out that the grass samples were taken from the sites subjected to the remediation activities performed according to recommendations of the UNEP experts as well as the experts from Bosnia and Herzegovina. Taking into consideration the high measurement uncertainties of the results for $^{238}$U in grass and hay, it was impossible to make further quantitative analysis on the presence or absence of depleted uranium in the observed samples. The lowest values of investigated radionuclides, with the exception of $^{40}$K, were recorded on the locality of Vitovlje village at the mountain Vlašić (Table 4). Obtained results were within the literature values for the areas with average background radioactivity (21). Statistical analysis of the results showed low values of standard deviation except for $^{40}$K (SD = 153.8). Statistically significant positive correlation between the activities of $^{238}$U and $^{226}$Ra (R=0.90) was indicative of the natural origin of uranium series radionuclides.

Obtained results did not point out at the significant increase in natural radionuclide amounts in the samples of rough-age feed. Slightly increased amounts of radionuclides of uranium and thorium series compared with the control locality, were due to the naturally high background activities in soil (Livno), presence of the ash from the coal power plant (Kakanj) or possible presence of very low concentrations of depleted uranium at sites targeted during the NATO strikes. Recorded activity concentrations in the samples of roughage feed indicated the acceptable levels of natural radionuclides in the diet of ruminants in the areas investigated, having in mind generally low bioavailability of observed radionuclides.
References


Konzentracije aktivnosti prirodnih radionuklida u sijenu i travi određenih područja Bosne i Hercegovine

Sažetak

Uvod i ciljevi
Područja visokog prirodnog backgrounda sa visokim sadržajima prirodnih radionuklida u obrokima predstavljaju posebnu zahvalnost. Ova rješenja zahtijevaju pouzdan određivanje radionuklida koji se nalaze u obroku. Ovim radom se zahtijeva preispitivanje koncentracija aktivnosti prirodnih radionuklida u sijenu i travi iz određenih područja Bosne i Hercegovine.

Materijal i metode
Uzorci trave i sijena iz određenih područja su izvorno ispitivani na visokim sadržajima prirodnih radionuklida sa posebnim nezglinima vezanim za područja Bosne i Hercegovine.

Rezultati i interpretacija
Ovisno o području različite su koncentracije aktivnosti prirodnih radionuklida. Od koncentracija 0,1 do 0,7 Bq kg⁻¹ za 226Ra i 0,06 do 0,7 Bq kg⁻¹ za 238U su na cvrčku više uzorci s visokim sadržajem radioaktivnosti. Najviše radioaktivnosti iznosi 40K na području Bosne i Hercegovine.

Zaključak
Zabilježene koncentracije aktivnosti prirodnih radionuklida su visoke u prvenstveno uzorkovanim obroku. Zabilježene su i posebne izvesnosti vezane za područja Bosne i Hercegovine.