

Concentration and characteristics of depleted uranium in water, air and biological samples collected in Serbia and Montenegro

Guogang Jia*, Maria Belli, Umberto Sansone, Silvia Rosamilia, Stefania Gaudino

Italian Environmental Protection Agency and Technical Services, Via V. Brancati 48, 00144 Roma, Italy

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Abstract

During the Balkan conflicts, in 1995 and 1999, depleted uranium (DU) rounds were employed and were left in the battlefield. Health concern is related to the risk arising from contamination of the environment with DU penetrators and dust. In order to evaluate the impact of DU on the environment and population in Serbia and Montenegro, radiological surveys of DU in water, air and biological samples were carried out over the period 27 October–5 November 2001. The uranium isotopic concentrations in biological samples collected in Serbia and Montenegro, mainly lichens and barks, were found to be in the range of $0.67\text{--}704\text{ Bq kg}^{-1}$ for ^{238}U , $0.48\text{--}93.9\text{ Bq kg}^{-1}$ for ^{234}U and $0.02\text{--}12.2\text{ Bq kg}^{-1}$ for ^{235}U , showing uranium levels to be higher than in the samples collected at the control sites. Moreover, ^{236}U was detectable in some of the samples. The isotopic ratios of $^{234}\text{U}/^{238}\text{U}$ showed DU to be detectable in many biological samples at all examined sites, especially in Montenegro, indicating widespread ground-surface DU contamination, albeit at very low level. The uranium isotopic concentrations in air obtained from the air filter samples collected in Serbia and Montenegro were found to be in the range of $1.99\text{--}42.1\text{ }\mu\text{Bq m}^{-3}$ for ^{238}U , $0.96\text{--}38.0\text{ }\mu\text{Bq m}^{-3}$ for ^{234}U , and $0.05\text{--}1.83\text{ }\mu\text{Bq m}^{-3}$ for ^{235}U , being in the typical range of natural uranium values. Thus said, most of the air samples are DU positive, this fact agreeing well with the widespread DU contamination detected in the biological samples. The uranium concentrations in water samples collected in Serbia and Montenegro were found to be in the range of $0.40\text{--}21.9\text{ mBq l}^{-1}$ for ^{238}U , $0.27\text{--}28.1\text{ mBq l}^{-1}$ for ^{234}U , and $0.01\text{--}0.88\text{ mBq l}^{-1}$ for ^{235}U , these values being much lower than those in mineral water found in central Italy and below the WHO guideline for drinking water. From a radiotoxicological point of view, at this moment there is no significant radiological risk related to these investigated sites in terms of possible DU contamination of water, air and/or plants.

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1. Introduction

Depleted uranium (DU) is the by-product of the nuclear industrial process that is used to enrich natural uranium ore for use in nuclear reactors and nuclear weapons. It is estimated that at present, approximately 6×10^5 tons of DU have been amassed in the USA

*Corresponding author. Tel.: +39 0650073219;
+39 0650073287.

E-mail address: jia@apat.it (G. Jia).

alone. DU is an extremely dense (19.05 g cm^{-3}), hard, autopyrophoric, relatively cheap metal, making it ideal for military application in armour piercing munitions and enhanced armour protection. DU was first extensively used by US Forces during the Gulf War in Operation Desert Storm in 1991, and then in the Balkan conflicts during 1995–1999. It has been reported that as a result of military use, the inventory of DU dispersed into the environment was about 320 tons in Iraq and Kuwait during the Gulf War in 1991, about 3 tons in Bosnia and Herzegovina in 1995, about 1 ton in Serbia and Montenegro and about 11 tons in Kosovo in 1999 (Hamilton, 2001; Bleise et al., 2003). Most recently, during the Iraq War in March and April 2003, >30 tons of DU was released into the environment. DU is a radioactive heavy metal that emits ionizing radiation of three types: alpha, beta and gamma, due to the decay of the radionuclides in the parent material, the various progeny and fission and/or activation products. Enhanced public awareness with regard to the health hazard arising from exposure to ionizing radiation in the environment contaminated by DU has led to increased efforts by the scientific community to assess the true effects of DU usage. In particular, the military use of DU has been a subject of considerable concern in the field of radiation protection, environmental protection and radioecology over the past fifteen years. As such, the United Nations Environment Programme (UNEP) has organized several missions, with participation from experts from inter-governmental agencies, well-known institutions and other interested parties, to conduct an overall assessment of the consequences of DU, post-conflict, on the environment and human settlements.

Following the Balkan conflict in 1999, during the period 5–19 November 2000, the Italian Environmental Protection Agency (ANPA) participated in the field mission to Kosovo. During the mission, water, vegetation, soil and smear samples were collected, the relevant data being published elsewhere (Sansone et al., 2001; UNEP, 2001; Jia et al., 2004a). Although DU has about 60% of the radioactivity of the natural uranium for an equal mass of substance (Cantaluppi and Degeto, 2000), elevated uranium activity concentrations have been observed in most of the surface soil samples and all of the biological samples collected in Kosovo. In some soils, elevations of even seven thousand times higher than the background uranium level have been observed. Subsequently, during the period 27 October–5 November 2001, the ANPA participated in the UNEP-coordinated field mission to Serbia and Montenegro (UNEP, 2002). During the mission, collection was primarily made of water, air [collected by the Greek Atomic Energy Commission (GAEC) and analysed by ANPA] and biological matrices (lichen, bark, etc.). In this current paper, the uranium activity concentrations in the collected samples are reported in detail, compar-

ison being made with samples uncontaminated by DU, collected at control sites in Italy. These data are of interest, serving as basic information for the particular affected sites, the quantity and quality of DU used in the conflict, and evaluation of the potential effects of DU on human health and/or the environment.

2. Materials and methods

2.1. Apparatus and reagents

The uranium sources were counted by alpha-spectrometry (Model 7401 with passivated implanted planar silicon detector and Genie-2000 spectroscopy system, Canberra, USA) with a counting efficiency of 31.2% and a background of $\leq 2 \times 10^{-6} \text{ s}^{-1}$ in the energy region of interest. The electrodeposition apparatus (model PL320QMD; Thurlby Thandar Instruments Ltd., England) was used to accumulate uranium in Perspex cells of 25 mm internal diameter on stainless-steel disks of 20 mm diameter in the medium of 0.025 M di-ammonium oxalate (Jia et al., 2002, 2004b). Chromatographic columns were 150 mm long and 9 mm internal diameter.

Standard solutions of ^{232}U and/or ^{236}U , Microthene (microporous polyethylene, 60–140 mesh), and tri-octylphosphine oxide (TOPO, 99%) were supplied by Amersham (G. B.), Ashland (Italy), and Fluka (Switzerland) respectively. FeCl_3 was used to prepare the carrier solution for uranium separation in water samples and all other reagents were analytical grade (Merck, Germany).

2.2. Column preparation

A solution (50 ml) of 0.3 M TOPO in cyclohexane was added to 50 g of Microthene; the mixture was stirred for several minutes until homogeneous and was then evaporated to eliminate cyclohexane at 50°C . The porous powder thus obtained contained about 10.4% TOPO. A portion (1.6 g) of the Microthene-TOPO powder, mixed with 3 ml concentrated HCl and some water, was transferred to a chromatographic column; after conditioning with 30 ml of 2 M HNO_3 , the column was ready for use.

2.3. Sampling sites

The criteria for selecting the sampling sites were that: (1) they should be locations for which the approximate number of DU rounds was known; (2) they should represent a range of environmental conditions and properties, and; (3) the areas to be examined should be safe from mines and unexploded ordnance. In fact, since the end of the Kosovo military conflict great efforts have been made by Serbian and Montenegrin authorities and experts to localize, measure and remove contamination

from DU and DU penetrators on ground. Eleven contaminated sites in Serbia had been surveyed up to October 2001 and, of these, five including Pljackovica, Borovac, Bratoselce, Bukurevac, Reljan, were investigated by the UNEP team. In Montenegro, one site (Cape Arza) partly surveyed by Montenegrin experts was also investigated by the UNEP team. The sampling code, date and geographical coordinates (UTM) are shown in Tables 1–3.

2.4. Sampling and sample preparation

In order to search for the presence of DU in botanical samples, at each investigated site lichen samples were collected using scalpels to obtain the material from mature tree trunks, the latter being as much as possible in the vertical position. Due to the lack of information concerning the prevailing wind direction, lichen samples were collected at different heights and positions along the trunks in order to make a composite sample representative of the different exposures. Bark samples were also collected at the same sites in order to investigate the relationship between the uranium content of bark samples and of lichen samples. Where available, lichen growing on rocks or soil, and moss samples were also collected. To minimize effects of cross-contamination following collection, all botanical samples were separately cleaned by removing visible soil particles and foreign bodies, classified into different species, dried at 105 °C, and ground and homogenized in a ceramic miller.

At sites in Serbia and Montenegro investigated by the UNEP team all air samples were collected by the team (UNEP, 2002). To measure low concentrations of radionuclides in air, sampling systems (GMWL-2000) equipped with high flow rate air pumps were used. At each of the sites (except Pljackovica) a filter system was placed in the centre of the fenced areas, while the other two sampling systems were placed in a symmetric position near the borders of the fenced areas. The three systems were arranged in a line parallel to the wind direction. The distance between the systems were 30–50 m. At Pljackovica, only two systems were operated and the filter systems were standing within 3 m from each other. At Cape Arza the fenced area was not easily accessed so the systems were placed outside the fence, near the neighboring decontamination area. During the investigation, the weather at the sites was warm and dry. The wind speed was low to moderate, except at the Reljan site where a strong wind prevailed.

In order to verify the presence of DU in drinking water, water samples from private wells and taps were collected and one seawater sample was also collected from the Cape Arza site. One litre of water was sampled at each site using a polyethylene bottle. Immediately after sampling, without any filtering of the water, the

pHs of all water samples were adjusted to <2 by adding HNO₃ to ensure that trace elements were kept in solution and to inhibit biological growth.

In order to evaluate the potential radiological impact of DU on the environment of Serbia and Montenegro, a number of environmental samples that were known to be free of contamination by DU, including water, tree trunk lichen and moss, and soil samples were also collected from Italy (Roma, Urbino) at the control sites (Tables 4–6). Detailed information about the control sites can be found elsewhere (Jia et al., 1997, 2002).

2.5. Method

As shown in Fig. 1, the radioanalytical procedure for determination of uranium isotopes in water, lichen, and soil samples mainly includes steps of sample pretreatment, leaching, uranium separation by a Microthene-TOPO column, electrodeposition and measurement by alpha-spectrometry. For more detailed procedure, please refer to the literature (Jia et al., 2002, 2004b).

In order to evaluate the reliability of the method, five reference or certified materials (IAEA-135 Sediment, IAEA-315 Sediment, IAEA-326 Soil, IAEA-327 Soil and IAEA-368 Sediment) have been analysed and the obtained results are all within the 95% confidence interval for the recommended or information values. The lower limits of detection of the method are 0.37 Bq kg⁻¹ (soil) and 0.22 mBq l⁻¹ (water) for ²³⁸U and ²³⁴U, 0.038 Bq kg⁻¹ (soil) and 0.022 mBq l⁻¹ (water) for ²³⁵U and ²³⁶U if 0.5 g of soil and 1 l of water are analysed. The average uranium yields for waters, lichens and soils are 74.5 ± 9.0%, 77.8 ± 4.9% and 89.4 ± 9.7%, respectively.

2.6. Calculation of DU fraction in possible DU contaminated environmental samples

Quantitative determination of the DU fraction (F_{DU}) in DU contaminated environmental samples is difficult, primarily because of the relatively large content of natural uranium present in the environment, with typical activity values in the range from 10 to 30 Bq kg⁻¹ for ²³⁸U or ²³⁴U. When background information of the uranium isotopic abundance is available, the presence of DU in samples can be assessed by variation of uranium isotopic ratios, such as ²³⁴U/²³⁸U and ²³⁵U/²³⁸U. To this end accurate measurement methods must be adopted which need to be validated very carefully prior to their use (Magnoni et al., 2001).

Uranium is present in nature as three isotopes, ²³⁸U, ²³⁴U and ²³⁵U. The ²³⁸U and ²³⁴U belong to the same decay chain (²³⁸U series), while the ²³⁵U belongs to the so-called actinium series, which is much less abundant. If not disturbed by any selective chemical effects (different leaching rate) or physical effects (alpha recoil)

Table 1
 sampling sites of lichens (L, Lr, Ls), barks (B), mosses (M), mushroom (F) and leaves (Le) in Serbia and Montenegro in 2001^a

Sample code	Sample type	Substrate	Sampling date	Sampling site	Coordinates, (UTM)	Sample code	Sample type	Substrate	Sampling date	Sampling site	Coordinates, (UTM)
L 1-3	Lichen	Tree	30-10-2001	Pljackovica	573657 714594	L 22-1a	Lichen	Tree	31-10-2001	Bratoselce	562282 688396
B 1-3	Bark	Tree	30-10-2001	Pljackovica	573657 714594	L 22-1b	Lichen	Tree	31-10-2001	Bratoselce	562282 688396
L 1-4a	Lichen	Tree	30-10-2001	Pljackovica	573657 714594	L 25-2a	Lichen	Tree	1-11-2001	Bukurevac	558156 688423
B 1-4a	Bark	Tree	30-10-2001	Pljackovica	573657 714594	L 25-2b	Lichen	Tree	1-11-2001	Bukurevac	558156 688423
L 2-3,4	Lichen	Tree	30-10-2001	Pljackovica	573664 714632	L 26-1a	Lichen	Tree	1-11-2001	Bukurevac	558084 688527
L 2-4a	Lichen	Tree	30-10-2001	Pljackovica	573664 714632	B 26-1a	Bark	Tree	1-11-2001	Bukurevac	558084 688527
L 2-7	Lichen	Tree	30-10-2001	Pljackovica	573664 714632	L 28-1a	Lichen	Tree	1-11-2001	Reljan	563208 685137
L 2-8	Lichen	Tree	30-10-2001	Pljackovica	573664 714632	L 29-1a	Lichen	Tree	1-11-2001	Reljan	563173 685079
B 2-8	Bark	Tree	30-10-2001	Pljackovica	573664 714632	B 29-1a	Bark	Tree	1-11-2001	Reljan	563173 685079
L 3-1	Lichen	Tree	30-10-2001	Pljackovica	573689 714660	Ls 30-1a	Lichen	Soil	1-11-2001	Reljan	563171 685063
L 3-2a	Lichen	Tree	30-10-2001	Pljackovica	573689 714660	Ls 30-1b	Lichen	Soil	1-11-2001	Reljan	563171 685063
L 3-2b	Lichen	Tree	30-10-2001	Pljackovica	573689 714660	Lr 31-1a	Lichen	Rock	1-11-2001	Reljan	563123 685073
L 3-2c	Lichen	Tree	30-10-2001	Pljackovica	573689 714660	M 31-2	Moss	Rock	1-11-2001	Reljan	563123 685073
L 3-3a	Lichen	Tree	30-10-2001	Pljackovica	573689 714660	L m1-1a	Lichen	Tree	3-11-2001	Cape Arza	300180 696077
L 3-3b	Lichen	Tree	30-10-2001	Pljackovica	573689 714660	L m2-1a	Lichen	Tree	3-11-2001	Cape Arza	300180 696077
L 3-6a	Lichen	Tree	30-10-2001	Pljackovica	573689 714660	Le m2-2	Leaf	Tree	1-11-2001	Reljan	563123 685073
L 3-6b	Lichen	Tree	30-10-2001	Pljackovica	573689 714660	L m3-1a	Lichen	Tree	3-11-2001	Cape Arza	300187 696137
Le 3-7	Leaf	Tree	30-10-2001	Pljackovica	573637 714629	B m3-1a	Bark	Tree	3-11-2001	Cape Arza	300187 696137
Lr 4-1	Lichen	Rock	30-10-2001	Pljackovica	573637 714629	Le m3-2	Leaf	Tree	3-11-2001	Cape Arza	300187 696137
L 5-1	Lichen	Tree	30-10-2001	Pljackovica	573637 714562	M m4-1	Moss	Rock	3-11-2001	Cape Arza	300188 696136
L 9-1a	Lichen	Tree	31-10-2001	Borovac	561910 693899	Le m5-1	Leaf	Tree	3-11-2001	Cape Arza	300188 696136
L 9-1c	Lichen	Tree	31-10-2001	Borovac	561910 693899	L m5-2a	Lichen	Tree	3-11-2001	Cape Arza	300257 696122
L 10-1a	Lichen	Tree	31-10-2001	Borovac	561894 693852	L m5-2b	Lichen	Tree	3-11-2001	Cape Arza	300257 696122
L 10-4a	Lichen	Tree	31-10-2001	Borovac	561894 693852	L m6-1a	Lichen	Tree	3-11-2001	Cape Arza	300065 696128
F 10-7	Mushroom	—	31-10-2001	Borovac	561838 694023	L m6-2a	Lichen	Tree	3-11-2001	Cape Arza	300065 696128
F 11-1	Mushroom	—	31-10-2001	Borovac	561838 694023	B m6-2a	Bark	Tree	3-11-2001	Cape Arza	300065 696128
L 12-1a	Lichen	Tree	31-10-2001	Borovac	561838 694023	Le m6-4	Leaf	Tree	3-11-2001	Cape Arza	300222 696092
L 12-1c	Lichen	Tree	31-10-2001	Borovac	561838 694023	L m7-1a	Lichen	Tree	3-11-2001	Cape Arza	300222 696092
L 12-2a	Lichen	Tree	31-10-2001	Borovac	561838 694023	L m7-1b	Lichen	Tree	3-11-2001	Cape Arza	300222 696092
L 12-2b	Lichen	Tree	31-10-2001	Borovac	561838 694023	L m7-1c	Lichen	Tree	3-11-2001	Cape Arza	300222 696092
L 12-2c	Lichen	Tree	31-10-2001	Borovac	561838 694023	Le m7-2	Leaf	Tree	3-11-2001	Cape Arza	300222 696092
Ls 18-1	Lichen	Soil	31-10-2001	Bratoselce	562274 688318	Le m7-3	Leaf	Tree	3-11-2001	Cape Arza	300234 696084
L 19-1a	Lichen	Tree	31-10-2001	Bratoselce	562305 688342	L m8-1	Lichen	Tree	3-11-2001	Cape Arza	300222 696092
L 20-1a	Lichen	Tree	31-10-2001	Bratoselce	562305 688312	B m8-1	Bark	Tree	3-11-2001	Cape Arza	300222 696092
L 21-2a	Lichen	Tree	31-10-2001	Bratoselce	562258 688363	Le m8-2a	Leaf	Tree	3-11-2001	Cape Arza	300222 696092
B 21-2a	Bark	Tree	31-10-2001	Bratoselce	562258 688363	Le m8-2b	Leaf	Tree	3-11-2001	Cape Arza	300222 696092
L 21-3a	Lichen	Tree	31-10-2001	Bratoselce	562258 688363	L m9-1a	Lichen	Tree	3-11-2001	Cape Arza	300234 696084
B 21-3a	Bark	Tree	31-10-2001	Bratoselce	562258 688363	B m9-1a	Bark	Tree	3-11-2001	Cape Arza	300234 696084
L 21-4	Lichen	Tree	31-10-2001	Bratoselce	562258 688363	Ls m10-1a	Lichen	Soil	3-11-2001	Cape Arza	300222 696092
B 21-4	Bark	Tree	31-10-2001	Bratoselce	562258 688363	L m11-1	Lichen	Tree	3-11-2001	Cape Arza	300222 696092

^aLr and Ls are lichens collected from rock and soil, respectively

Table 2
Sampling sites of air in Serbia and Montenegro in 2001 (AF: Air filter)

Sample code	Sampling date	Sampling site	Sampling volume (m ³)	Coordinates (UTM)	
AF 1-1	30-10-2001	Pljackovica	156	573613	714596
AF 1-3	30-10-2001	Pljackovica	156	573613	714596
AF 2-1	31-10-2001	Borovac	150	561956	694025
AF 2-2	31-10-2001	Borovac	150	561969	694064
AF 2-3	31-10-2001	Borovac	150	561935	693979
AF 3-1	31-10-2001	Bratoselce	116	562271	688310
AF 3-2	31-10-2001	Bratoselce	116	562278	688285
AF 3-3	31-10-2001	Bratoselce	116	562292	688251
AF 4-1	1-11-2001	Bukurevac	130	558190	688787
AF 4-2	1-11-2001	Bukurevac	130	558209	688731
AF 4-3	1-11-2001	Bukurevac	130	558165	688761
AF 5-1	1-11-2001	Reljan	130	553168	685120
AF 5-2	1-11-2001	Reljan	130	553155	685074
AF 5-3	1-11-2001	Reljan	130	553150	685032
AF 6-1	3-11-2001	Cape Arza	192	300146	696097
AF 6-2	3-11-2001	Cape Arza	192	300119	696100
AF 6-3	3-11-2001	Cape Arza	192	300174	696120

Table 3
Sampling sites of tap water (TW), well water (WW) and sea water (SW) in Serbia and Montenegro in 2001

Sample code	Sampling date	Sampling site	Coordinates (UTM)	
TW6	30-10-2001	Pržar	574041	713641
WW7	30-10-2001	Pržar	573796	713729
TW8	30-10-2001	Pržar (Hotel)	573707	713478
TW13	31-10-2001	Borovac	561577	695276
WW14	31-10-2001	Borovac	561162	694635
WW15	31-10-2001	Borovac	562061	694078
WW16	31-10-2001	Borovac	562087	694072
WW17	31-10-2001	Borovac	562028	694185
WW23	31-10-2001	Bratoselce	—	—
WW27	1-11-2001	Bukurevac	558184	688800
SW12	3-11-2001	Cape Arza	300239	696157

they should be in radioactive equilibrium. In the case of equilibrium, the isotopic abundances of natural uranium by weight are 99.2745% of ²³⁸U, 0.00550% of ²³⁴U and 0.720% of ²³⁵U with a ²³⁴U/²³⁸U mass ratio ($R_{m234/238}$) of 5.54×10^{-5} and a ²³⁵U/²³⁸U mass ratio ($R_{m235/238}$) of 0.00725. If the corresponding activity ratio is considered, the ²³⁴U/²³⁸U value ($R_{a234/238}$) is about 1.00, and the ²³⁵U/²³⁸U value ($R_{a235/238}$) is about 0.0466 (Invanovich and Harmon, 1992). Thus said, it is a matter of fact that the selective chemical and physical effects always occur in biological and environmental samples, to some greater or lesser degree.

In order to estimate quantitatively the DU fraction in lichen, air and water samples collected in Serbia and Montenegro, basic information about the uranium isotopic abundances in uncontaminated environmental

samples should also be investigated. For this purpose and as already mentioned, some lichen, moss, soil and water samples uncontaminated from DU have been collected from central Italy and analysed for the knowledge of the variation of their natural uranium isotopic abundances. Soil samples were also selected, as the manifest ²³⁴U/²³⁸U ratio variation in air at a given place will also reflect to an extent the ratio variation in soil at that location.

The obtained activity concentrations of uranium isotopes in lichen, moss and soil samples uncontaminated from DU are given in Tables 4 and 5, showing that the average ²³⁴U/²³⁸U activity ratios are 1.006 ± 0.097 for lichen, 1.027 ± 0.082 for moss and 0.993 ± 0.033 for soil. The obtained isotopic abundances by weight are $(99.05 \pm 0.43)\%$ of ²³⁸U, $(0.00539 \pm$

Table 4

^{238}U , ^{234}U and ^{235}U concentrations (in Bq kg^{-1}) in tree trunk lichen (L) and moss (M) samples collected in central Italy (Urbino) in 1997

Sample code	Sample weight, g	U yield, %	^{238}U	^{234}U	^{235}U	$^{234}\text{U}/^{238}\text{U}$	$^{235}\text{U}/^{238}\text{U}$
L1	1.2107	82.3	2.48 ± 0.16	2.50 ± 0.16	0.10 ± 0.03	1.006 ± 0.091	0.040 ± 0.014
L3	1.0394	66.3	2.71 ± 0.22	2.53 ± 0.21	0.23 ± 0.07	0.931 ± 0.107	0.086 ± 0.026
L4	1.1837	89.9	4.67 ± 0.23	4.61 ± 0.23	0.23 ± 0.05	0.988 ± 0.070	0.049 ± 0.011
L6	1.1074	85.8	2.56 ± 0.18	2.58 ± 0.19	0.04 ± 0.03	1.008 ± 0.103	0.017 ± 0.012
L7	1.0363	76.4	2.54 ± 0.14	2.69 ± 0.14	0.19 ± 0.04	1.056 ± 0.137	0.073 ± 0.031
L9	1.0277	73.0	2.21 ± 0.17	2.39 ± 0.18	0.12 ± 0.04	1.082 ± 0.115	0.053 ± 0.020
L10	1.0625	52.0	2.96 ± 0.22	2.98 ± 0.23	0.28 ± 0.07	1.009 ± 0.108	0.094 ± 0.026
L11	1.0481	71.1	1.46 ± 0.16	1.27 ± 0.15	0.08 ± 0.04	0.869 ± 0.140	0.052 ± 0.031
L21	1.0702	74.7	4.68 ± 0.32	5.17 ± 0.34	0.19 ± 0.07	1.105 ± 0.106	0.040 ± 0.015
L22	1.0873	78.7	1.37 ± 0.13	1.40 ± 0.13	0.05 ± 0.03	1.021 ± 0.136	0.035 ± 0.022
L23	1.1074	70.2	2.91 ± 0.21	2.44 ± 0.19	0.09 ± 0.04	0.840 ± 0.090	0.032 ± 0.015
L24	1.1638	78.0	1.01 ± 0.10	0.85 ± 0.10	0.04 ± 0.03	0.842 ± 0.130	0.042 ± 0.027
L25	1.0126	78.0	2.33 ± 0.17	2.25 ± 0.17	0.15 ± 0.05	0.963 ± 0.100	0.064 ± 0.021
L26	1.1525	80.3	1.92 ± 0.14	1.92 ± 0.14	0.10 ± 0.03	1.000 ± 0.099	0.051 ± 0.018
L27	1.0681	84.5	3.81 ± 0.22	3.99 ± 0.22	0.27 ± 0.06	1.047 ± 0.107	0.070 ± 0.023
L28	1.1423	65.3	1.70 ± 0.19	1.98 ± 0.20	0.17 ± 0.07	1.161 ± 0.177	0.101 ± 0.041
L29	1.0363	78.5	2.17 ± 0.19	2.53 ± 0.20	0.22 ± 0.06	1.168 ± 0.137	0.101 ± 0.031
Mean or range	1.0126–1.2107	75.6 ± 9.0	1.01–4.68	0.85–5.17	0.04–0.28	1.006 ± 0.097	0.059 ± 0.025
M1	1.0729	69.9	2.91 ± 0.26	3.30 ± 0.28	0.23 ± 0.08	1.132 ± 0.139	0.081 ± 0.028
M2	1.0141	75.3	1.53 ± 0.16	1.80 ± 0.17	0.09 ± 0.05	1.180 ± 0.167	0.060 ± 0.030
M3	1.0589	78.0	5.13 ± 0.35	4.67 ± 0.33	0.20 ± 0.07	0.910 ± 0.089	0.038 ± 0.014
M4	1.0301	71.8	3.55 ± 0.25	3.81 ± 0.26	0.31 ± 0.08	1.072 ± 0.106	0.086 ± 0.023
M5	1.3123	75.0	1.62 ± 0.12	1.77 ± 0.12	0.06 ± 0.03	1.090 ± 0.110	0.040 ± 0.017
M6	1.0203	77.0	5.33 ± 0.35	5.56 ± 0.36	0.31 ± 0.09	1.043 ± 0.096	0.059 ± 0.017
M10	1.0007	75.6	1.37 ± 0.12	1.33 ± 0.14	0.06 ± 0.03	0.972 ± 0.133	0.046 ± 0.024
M20	1.0870	82.3	1.50 ± 0.12	1.53 ± 0.12	0.17 ± 0.04	1.019 ± 0.113	0.116 ± 0.030
M21	1.2246	91.2	6.90 ± 0.37	6.19 ± 0.34	0.27 ± 0.07	0.897 ± 0.069	0.039 ± 0.010
M22	1.0990	97.0	6.25 ± 0.34	6.17 ± 0.34	0.27 ± 0.07	0.988 ± 0.077	0.044 ± 0.011
M23	1.0984	73.7	1.18 ± 0.14	1.15 ± 0.14	0.06 ± 0.04	0.978 ± 0.167	0.049 ± 0.033
M26	1.1952	89.0	7.14 ± 0.38	7.69 ± 0.40	0.31 ± 0.07	1.076 ± 0.080	0.044 ± 0.011
M27	1.0314	81.7	2.89 ± 0.19	2.89 ± 0.19	0.11 ± 0.04	1.000 ± 0.093	0.038 ± 0.014
Mean or range	1.0007–1.3123	79.8 ± 8.1	1.18–7.14	1.15–7.69	0.06–0.31	1.027 ± 0.082	0.057 ± 0.024

0.00057)% of ^{234}U , (0.943 ± 0.430)% of ^{235}U [$R_{m234/238}$: (5.45 ± 0.59) $\times 10^{-5}$; $R_{m235/238}$: 0.00954 ± 0.00439] in lichen, (99.12 ± 0.36)% of ^{238}U , (0.00548 ± 0.00043)% of ^{234}U , (0.873 ± 0.036)% of ^{235}U [$R_{m234/238}$: (5.53 ± 0.44) $\times 10^{-5}$; $R_{m235/238}$: 0.00882 ± 0.00369] in moss and (99.22 ± 0.082)% of ^{238}U , (0.00530 ± 0.00018)% of ^{234}U , (0.780 ± 0.085)% of ^{235}U [$R_{m234/238}$: (5.34 ± 0.18) $\times 10^{-5}$; $R_{m235/238}$: 0.00786 ± 0.00087] in soil. These results show a similar characteristic to that of natural uranium in these samples.

Disequilibrium of ^{234}U with ^{238}U was observed in water samples (Table 6). The $^{234}\text{U}/^{238}\text{U}$ activity ratios are higher and variable in mineral or drinking waters (1.42 ± 0.19), and relatively constant in river waters (1.18 ± 0.04) and seawaters (1.15 ± 0.06). The observed isotopic abundances by weight are (99.20 ± 0.15)% of ^{238}U , (0.00757 ± 0.0010)% of ^{234}U , (0.788 ± 0.149)%

of ^{235}U [$R_{m234/238}$: (7.63 ± 1.01) $\times 10^{-5}$; $R_{m235/238}$: 0.00795 ± 0.00152] in drinking water, (99.28 ± 0.10)% of ^{238}U , (0.00634 ± 0.00023)% of ^{234}U , (0.718 ± 0.105)% of ^{235}U [$R_{m234/238}$: (6.86 ± 1.88) $\times 10^{-5}$; $R_{m235/238}$: 0.00733 ± 0.00206] in river water and (99.16 ± 0.11)% of ^{238}U , (0.00611 ± 0.00033)% of ^{234}U , (0.831 ± 0.113)% of ^{235}U [$R_{m234/238}$: (6.47 ± 1.88) $\times 10^{-5}$; $R_{m235/238}$: 0.00733 ± 0.00216] in seawater.

The uranium isotopic abundance (m/m) of DU were obtained by analysing smear samples obtained from the penetrators used in the Balkan conflict (Jia et al., 2004a), being (99.77 ± 0.01)% of ^{238}U , (0.000678 ± 0.000014)% of ^{234}U , (0.224 ± 0.009)% of ^{235}U , and (0.00295 ± 0.00020)% of ^{236}U ($R_{m234/238}$: 6.79×10^{-6} ; $R_{m235/238}$: 0.00224 ; $R_{m236/238}$: 2.96×10^{-5}).

Due to the fact that the activity concentrations of ^{234}U and ^{238}U can be accurately measured by the

Table 5
 ^{238}U , ^{234}U and ^{235}U concentrations (in Bq kg^{-1}) in soil samples collected in Italy in December 1998

Sample code	Sample weight (g)	U yield (%)	^{238}U	^{234}U	^{235}U	$^{234}\text{U}/^{238}\text{U}$	$^{235}\text{U}/^{238}\text{U}$
S1	0.5110	81.7	29.9 ± 1.3	31.3 ± 1.3	1.65 ± 0.24	1.048 ± 0.062	0.055 ± 0.008
S2	0.5298	88.9	30.9 ± 1.2	30.8 ± 1.2	1.65 ± 0.22	0.996 ± 0.056	0.053 ± 0.007
S3	0.5170	83.0	25.6 ± 1.1	26.4 ± 1.1	1.34 ± 0.21	1.033 ± 0.062	0.052 ± 0.009
S4	0.5080	100.7	31.9 ± 1.3	31.7 ± 1.3	1.58 ± 0.23	0.993 ± 0.056	0.049 ± 0.007
S5	0.5503	94.7	29.5 ± 1.1	28.6 ± 1.1	1.50 ± 0.20	0.969 ± 0.052	0.051 ± 0.007
S6	0.5358	95.2	29.3 ± 1.1	28.4 ± 1.1	1.42 ± 0.20	0.968 ± 0.053	0.048 ± 0.007
S7	0.5190	97.5	31.7 ± 1.2	31.8 ± 1.2	1.36 ± 0.19	1.003 ± 0.054	0.043 ± 0.006
S8	0.5078	92.9	29.1 ± 1.2	29.5 ± 1.2	1.28 ± 0.20	1.012 ± 0.058	0.044 ± 0.007
S9	0.4999	81.6	29.7 ± 1.2	29.6 ± 1.2	1.68 ± 0.24	0.997 ± 0.059	0.057 ± 0.008
S10	0.5224	87.9	29.8 ± 1.5	27.1 ± 1.4	1.52 ± 0.30	0.910 ± 0.066	0.051 ± 0.010
S11	0.5337	73.9	33.0 ± 1.2	33.3 ± 1.2	1.48 ± 0.19	1.009 ± 0.052	0.045 ± 0.006
S12	0.5100	80.3	28.9 ± 1.6	29.3 ± 1.6	1.18 ± 0.29	1.015 ± 0.079	0.041 ± 0.010
S13	0.5505	76.3	28.9 ± 1.2	28.4 ± 1.2	1.32 ± 0.21	0.983 ± 0.058	0.046 ± 0.007
S14	0.5293	99.0	53.1 ± 1.7	53.8 ± 1.7	2.48 ± 0.23	1.012 ± 0.046	0.047 ± 0.005
S15	0.5259	98.8	29.4 ± 1.0	30.1 ± 1.1	1.67 ± 0.19	1.025 ± 0.052	0.057 ± 0.007
S16	0.5148	97.2	29.5 ± 1.1	30.1 ± 1.1	1.56 ± 0.19	1.022 ± 0.052	0.053 ± 0.007
S17	0.5184	76.7	33.1 ± 1.4	30.3 ± 1.3	1.67 ± 0.25	0.916 ± 0.055	0.050 ± 0.008
S18	0.5578	86.2	27.2 ± 1.1	27.1 ± 1.1	1.32 ± 0.20	0.997 ± 0.058	0.049 ± 0.008
S20	0.5157	95.2	29.4 ± 1.2	29.2 ± 1.2	1.73 ± 0.23	0.994 ± 0.056	0.059 ± 0.008
S21	0.5207	89.9	30.8 ± 1.2	29.9 ± 1.2	1.36 ± 0.20	0.968 ± 0.054	0.044 ± 0.007
S22	0.5254	82.2	31.2 ± 1.3	31.3 ± 1.3	1.80 ± 0.24	1.003 ± 0.057	0.058 ± 0.008
S23	0.5027	96.1	29.8 ± 1.2	29.2 ± 1.2	1.80 ± 0.24	0.980 ± 0.056	0.060 ± 0.008
Mean or range	0.4990–0.5578	88.9 ± 8.4	25.6–53.1	26.4–53.8	1.18–2.48	0.993 ± 0.033	0.051 ± 0.006

Table 6
 ^{238}U , ^{234}U and ^{235}U concentrations (in mBq l^{-1}) in drinking water (DW), filtered river water (RW) and sea water (SW) samples collected in Italy

Sample code	Water volume (l)	U yield (%)	^{238}U	^{234}U	^{235}U	$^{234}\text{U}/^{238}\text{U}$	$^{235}\text{U}/^{238}\text{U}$
DW1	1.000	73.5	7.50 ± 0.39	7.84 ± 0.40	0.51 ± 0.10	1.05 ± 0.08	0.068 ± 0.013
DW2	1.000	62.6	17.7 ± 1.0	29.8 ± 1.5	0.63 ± 0.16	1.68 ± 0.13	0.036 ± 0.009
DW3	1.000	71.0	18.0 ± 0.8	27.2 ± 1.1	0.97 ± 0.14	1.52 ± 0.09	0.054 ± 0.008
DW4	1.000	81.7	97.5 ± 4.0	131 ± 5	4.82 ± 0.45	1.34 ± 0.08	0.049 ± 0.005
DW5	0.500	70.4	103 ± 4	135 ± 4	4.77 ± 0.43	1.31 ± 0.06	0.046 ± 0.004
DW6	0.500	62.0	33.9 ± 2.7	48.1 ± 3.4	2.15 ± 0.60	1.42 ± 0.15	0.063 ± 0.018
DW7	0.538	71.7	35.7 ± 1.5	48.4 ± 2.0	1.73 ± 0.26	1.36 ± 0.08	0.049 ± 0.008
DW8	1.000	90.9	0.30 ± 0.06	0.49 ± 0.07	0.02 ± 0.02	1.62 ± 0.38	0.049 ± 0.057
DW9	1.000	75.5	4.76 ± 0.30	7.03 ± 0.38	0.21 ± 0.06	1.48 ± 0.12	0.045 ± 0.014
Mean or range	0.500–1.000	73.3 ± 9.0	0.30–103	0.49–135	0.02–4.82	1.42 ± 0.19	0.051 ± 0.010
RW1	1.000	59.5	19.4 ± 1.0	23.7 ± 1.1	0.72 ± 0.16	1.22 ± 0.09	0.037 ± 0.008
RW2	0.500	77.0	29.4 ± 1.7	33.6 ± 1.8	1.49 ± 0.33	1.14 ± 0.09	0.051 ± 0.012
RW3	0.500	88.7	30.8 ± 1.6	37.7 ± 1.8	1.62 ± 0.30	1.22 ± 0.09	0.053 ± 0.010
RW4	0.500	80.0	22.8 ± 1.4	26.4 ± 1.5	1.04 ± 0.28	1.16 ± 0.10	0.046 ± 0.012
Mean or range	0.500–1.000	76.3 ± 12.2	19.4–30.8	23.7–37.7	0.72–1.62	1.18 ± 0.04	0.047 ± 0.007
SW1	0.500	88.8	40.2 ± 2.0	41.9 ± 2.0	2.62 ± 0.40	1.04 ± 0.07	0.065 ± 0.011
SW2	1.000	68.9	39.6 ± 1.9	45.8 ± 2.2	1.92 ± 0.29	1.16 ± 0.08	0.048 ± 0.008
SW3	1.000	68.0	42.5 ± 1.8	49.1 ± 2.0	2.41 ± 0.27	1.16 ± 0.07	0.057 ± 0.007
SW4	1.000	70.2	42.7 ± 1.7	51.5 ± 2.0	1.98 ± 0.22	1.21 ± 0.07	0.046 ± 0.006
SW5	1.000	83.0	42.4 ± 1.4	49.2 ± 1.6	2.25 ± 0.18	1.16 ± 0.05	0.053 ± 0.005
Mean or range	0.500–1.000	75.8 ± 9.5	41.5 ± 1.5	47.5 ± 3.7	2.24 ± 0.29	1.15 ± 0.06	0.054 ± 0.008

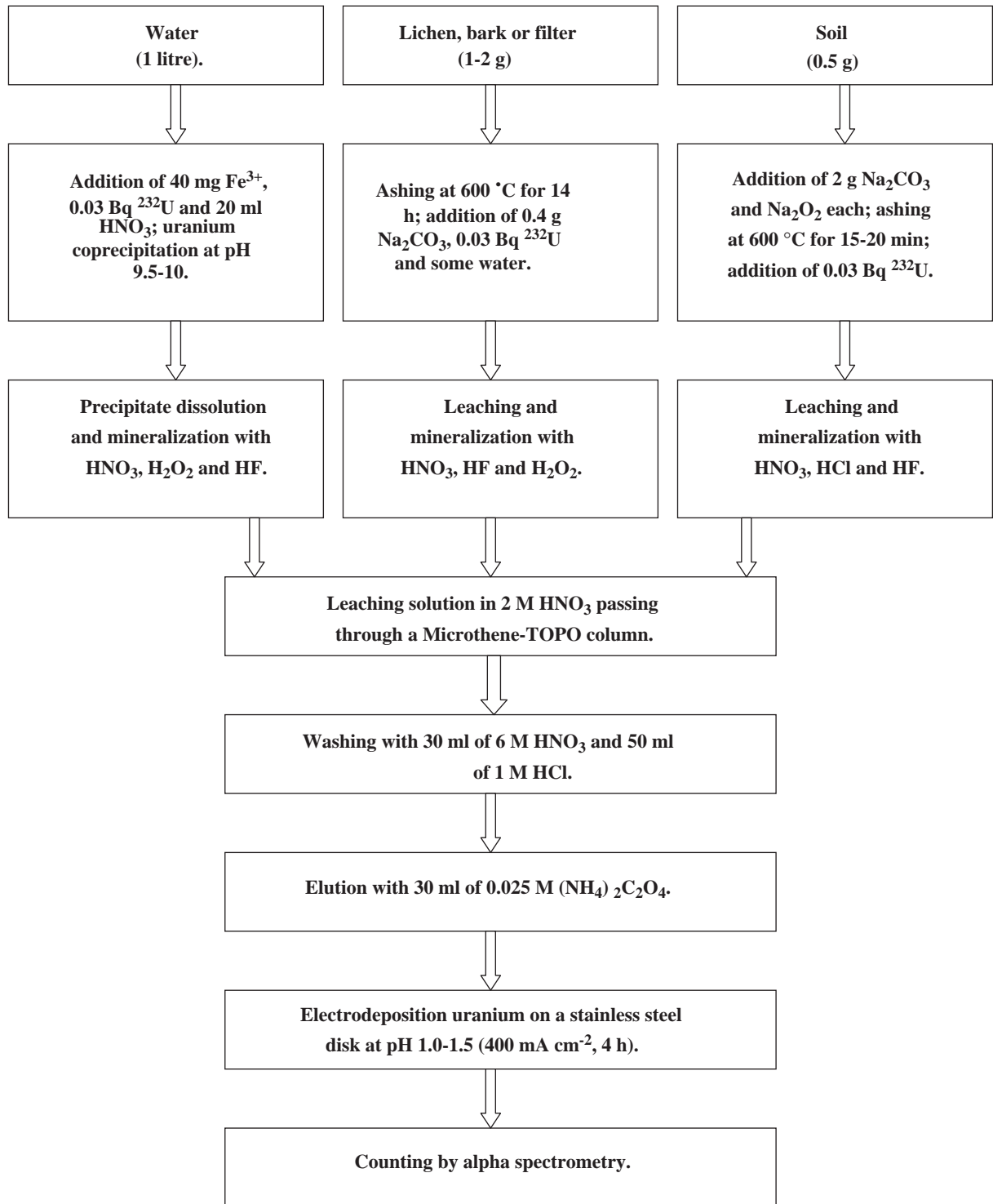


Fig. 1. Recommended procedure for determination of uranium in environmental samples by alpha-spectrometry.

extraction chromatographic/alpha-spectrometry method, activity concentrations of ²³⁴U and ²³⁸U obtained from the samples analyses have been used to calculate

the DU fraction in the lichen, moss, water and air (soil) samples. Assuming that in a sample with some DU the mass proportion of DU is F_{DU} , the natural uranium

represents the fraction $1 - F_{DU}$. Therefore, based on the average uranium isotopic abundances obtained by weight, which are 99.156% of ^{238}U and 0.00536% of ^{234}U in lichen, moss or air (soil) samples uncontaminated from DU, and 99.773% of ^{238}U and 0.000678% of ^{234}U in DU of the penetrators, the mass ratio between ^{234}U and ^{238}U ($R_{m234/238}$) in a lichen, moss or air (soil) sample is given by the formula

$$R_{m234/238} = \frac{0.00536(1 - F_{DU}) + 0.000678F_{DU}}{99.156(1 - F_{DU}) + 99.773F_{DU}}. \quad (1)$$

The activity ratio between ^{234}U and ^{238}U , $R_{a234/238}$ is given by $2.312 \times 10^5 R_{m234/238}/12.44$. For $F_{DU} = 0$, the $R_{a234/238}$ is 1.00; for $F_{DU} = 0.5$, the $R_{a234/238}$ is 0.564; and for $F_{DU} = 1$, the $R_{a234/238}$ is 0.126, etc. The $R_{m234/238}$ value can be obtained from the sample analyses, therefore, F_{DU} in lichen, moss and air (soil) samples collected in Serbia and Montenegro is calculated as

$$F_{DU} = \frac{0.00536 - 99.156R_{m234/238}}{0.00468 + 0.617R_{m234/238}}. \quad (2)$$

Due to the obvious radioactive disequilibrium between ^{234}U and ^{238}U in water samples, uranium isotopic abundances by weight in seawater, which are 99.162% of ^{238}U and 0.00611% of ^{234}U , are used to estimate the DU fraction in the drinking water samples collected in Serbia and Montenegro. $R_{m234/238}$ in water samples is given from the following equation:

$$R_{m234/238} = \frac{0.00611(1 - F_{DU}) + 0.000678F_{DU}}{99.162(1 - F_{DU}) + 99.773F_{DU}} \quad (3)$$

and F_{DU} in drinking water is

$$F_{DU} = \frac{0.00611 - 99.162R_{m234/238}}{0.00540 + 0.610R_{m234/238}}. \quad (4)$$

Experimental determination of the isotopic ratio between ^{234}U and ^{238}U is affected by uncertainties, the magnitude of which depends upon the accuracy and precision of the measurements of the single isotopes. It appears clear that the values of the isotopic ratios overlap with those of natural uranium when the DU fraction is not significant. Magnoni et al. (2001) have assessed the sensitivity of the technique, and concluded that the threshold for detection of the DU fraction in a typical soil or other matrices sample containing 5–60 Bq kg⁻¹ of natural ^{238}U , is about 10–20% of the total uranium present in the sample.

3. Results and discussion

The pyrophoric nature of DU is of special relevance to the environmental impacts resulting from its use in ordnance and armoured. The characteristics and behaviour of DU anti-armour rounds fired by A-10

aircraft in the environment can principally be classified into two scenarios. First, when rounds hit either non-armoured targets or miss targets, they will generally remain intact, passing through the target and/or becoming buried in the ground. The depth depends on the angle of the round, the speed of the plane, the type of target and the nature of the ground surface. In clay soils, the penetrators may reach more than 2 m in depth. In this scenario, there are risks of external exposure and underground water contamination due to the mobilization of DU in soil profile after corrosion and dissolution on the basis of the acidity and reducing properties of the environment and the hydrological characteristics of the region. Second, when rounds hit the armoured or hard targets, normally 10–35% (maximum of 70%) of the penetrators become aerosol on impact with armour and the DU dust catches fire (Rand, 1999). Most of the dust particles are < 5 μm in size and spread according to wind direction. The DU dust is black and consists mainly of uranium oxides. A target that has been hit by DU ammunition can be recognized by the black dust cover in and around the target (USAEPI., 1994). The dust formed during the penetration of armoured vehicles can be dispersed into the environment, contaminating the air, water, vegetation and soil. One opinion is that such contamination should be limited to within about 100 m of the target (CHPPM, 2000; Nellis, 1997), while another is that these uranium oxide particles can be dispersed with a radius of several kilometres (UNEP, 2001). It is obvious that the contamination range of DU largely depends on the local climate and meteorological conditions. After deposition, the small penetrator fragments and DU dust may also be redistributed due to: (1) the resuspension by wind; (2) transportation by insects, worms and some human activities (cultivation, irrigation and fertilization); (3) biological and chemical processes by corrosion or oxidation and reduction, and; (4) rainwater, surface water and underground water. It is a matter of record that the second scenario is responsible for most of the air, water, vegetation and soil contamination and can cause DU to become a primary radiation source (UNEP, 2001, 2002).

The obtained activity concentrations of uranium isotope in the samples collected in Serbia, Montenegro and also from the control sites are given in Tables 4–10. The reported uncertainty for individual analysis in the tables is 1σ , this being estimated from the uncertainties associated with the tracer activity, the addition of the tracer to the sample and the counting statistics of the sample and the blank, etc.

3.1. Uranium in lichen, moss, mushroom, bark and leaf samples

Lichens, mosses, mushrooms, barks and leaves have been widely used as sensitive, inexpensive and easily

Table 7

^{238}U , ^{234}U , ^{235}U and ^{236}U concentrations (in Bq kg^{-1}) in biological samples (L: lichen; B: bark; Le: leaf; F: mushroom; M: moss) collected in Serbia from 27 October to 5 November 2001

Site	Sample code	Lichen species	U yield (%)	^{238}U	^{234}U	^{235}U	^{236}U	$^{234}\text{U}/^{238}\text{U}$	$^{235}\text{U}/^{238}\text{U}$	$^{236}\text{U}/^{238}\text{U}$	F_{DU} , %
Pljackovica	L 1-3	Parmelia borrieri	66.7	4.04 ± 0.29	3.87 ± 0.28	0.25 ± 0.08	ND*	0.957 ± 0.098	0.061 ± 0.021	ND	≤ 5.3
	B 1-3	—	44.2	1.83 ± 0.15	2.04 ± 0.16	0.13 ± 0.05	ND	1.11 ± 0.12	0.069 ± 0.029	ND	ND
	L 1-4a	Parmelia borrieri	84.2	2.65 ± 0.26	2.51 ± 0.25	0.17 ± 0.08	ND	0.946 ± 0.134	0.063 ± 0.029	ND	≤ 6.6
	B 1-4a	—	43.7	1.88 ± 0.14	1.67 ± 0.14	0.05 ± 0.04	ND	0.884 ± 0.099	0.028 ± 0.021	ND	≤ 13.6
	L 2-3,4	Hypogymnia farinacea	74.4	3.22 ± 0.18	2.77 ± 0.16	0.16 ± 0.04	0.007 ± 0.011	0.862 ± 0.069	0.058 ± 0.013	0.0021 ± 0.0035	≤ 16.1
	L 2-4a	Parmelia acetabulum	73.8	3.57 ± 0.21	3.30 ± 0.20	0.16 ± 0.05	ND	0.926 ± 0.079	0.043 ± 0.015	ND	≤ 8.9
	L 2-7	Parmelia borrieri	66.8	2.49 ± 0.29	2.21 ± 0.27	0.08 ± 0.07	0.006 ± 0.037	0.889 ± 0.152	0.032 ± 0.029	0.0022 ± 0.0149	≤ 13.1
	L 2-8	Parmelia borrieri	93.7	1.73 ± 0.12	1.81 ± 0.12	0.10 ± 0.03	ND	1.04 ± 0.097	0.060 ± 0.020	ND	ND
	B 2-8	—	56.4	0.81 ± 0.08	0.82 ± 0.08	0.03 ± 0.03	ND	1.02 ± 0.15	0.037 ± 0.035	ND	ND
	L 3-1	Parmelia tiliacea	72.3	2.82 ± 0.29	3.46 ± 0.32	0.19 ± 0.09	ND	1.23 ± 0.17	0.068 ± 0.032	ND	ND
	L 3-1	Pseudevernia furfuracea	75.6	5.21 ± 0.23	5.12 ± 0.23	0.24 ± 0.04	ND	0.983 ± 0.062	0.046 ± 0.009	ND	ND
	B 3-1	—	93.7	5.92 ± 0.60	5.77 ± 0.59	0.16 ± 0.11	ND	0.976 ± 0.140	0.026 ± 0.020	ND	ND
	L 3-2a	Pseudevernia furfuracea	57.6	0.70 ± 0.08	0.69 ± 0.08	0.04 ± 0.03	ND	0.990 ± 0.167	0.061 ± 0.047	ND	ND
	L 3-2b	Hypogymnia farinacea	61.3	1.51 ± 0.11	1.42 ± 0.11	0.08 ± 0.03	ND	0.938 ± 0.100	0.050 ± 0.023	ND	≤ 7.5
	L 3-2b	Hypogymnia farinacea	72.8	3.15 ± 0.15	3.18 ± 0.15	0.17 ± 0.03	ND	1.01 ± 0.067	0.054 ± 0.010	ND	ND
	L 3-2c	Parmelia sulcata	67.0	4.76 ± 0.40	5.03 ± 0.41	0.30 ± 0.11	ND	1.06 ± 0.124	0.063 ± 0.023	ND	ND
	L 3-3a	Pseudevernia furfuracea	58.3	0.89 ± 0.13	0.89 ± 0.13	0.02 ± 0.04	ND	1.00 ± 0.206	0.027 ± 0.046	ND	ND
	L 3-3b	Hypogymnia farinacea	76.4	1.50 ± 0.16	1.02 ± 0.14	0.04 ± 0.04	0.004 ± 0.010	0.679 ± 0.118	0.046 ± 0.029	0.0026 ± 0.0177	36.9
	L 3-6a	Hypogymnia farinacea	68.3	1.48 ± 0.12	1.48 ± 0.12	0.04 ± 0.03	ND	1.00 ± 0.114	0.026 ± 0.021	ND	ND
	L 3-6b	Hypogymnia farinacea	78.0	0.85 ± 0.13	0.80 ± 0.12	0.12 ± 0.06	ND	0.942 ± 0.201	0.141 ± 0.071	ND	≤ 7.1
Le 3-7	—	71.6	0.22 ± 0.05	0.28 ± 0.05	0.02 ± 0.01	ND	1.32 ± 0.36	0.088 ± 0.065	ND	ND	
Lr 4-1	Lecanora muralis	103	9.63 ± 0.55	9.38 ± 0.54	0.69 ± 0.14	ND	0.974 ± 0.079	0.071 ± 0.015	ND	ND	
Lr 4-1	Lecanora muralis	70.6	13.1 ± 0.9	14.1 ± 1.0	1.02 ± 0.20	ND	1.08 ± 0.10	0.077 ± 0.016	ND	ND	
Lr 4-1	Lecanora muralis	96.4	9.46 ± 0.35	9.71 ± 0.36	0.52 ± 0.06	ND	1.03 ± 0.05	0.055 ± 0.007	ND	ND	
Lr 4-1	Lecanora muralis	98.3	13.6 ± 0.6	14.6 ± 0.6	0.64 ± 0.10	ND	1.08 ± 0.07	0.047 ± 0.007	ND	ND	
Lr 4-1	Lecanora muralis	97.7	20.3 ± 1.0	21.3 ± 1.0	1.30 ± 0.23	ND	1.05 ± 0.07	0.064 ± 0.012	ND	ND	
Lr 4-1	Lecanora muralis	101	13.8 ± 0.4	14.1 ± 0.4	0.68 ± 0.06	0.066 ± 0.017	1.02 ± 0.05	0.049 ± 0.005	0.0048 ± 0.0013	ND	
L 5-1	Ramalina farinacea	60.0	0.72 ± 0.12	0.49 ± 0.11	0.03 ± 0.05	ND	0.687 ± 0.192	0.037 ± 0.066	—	35.9	
Borovac	L 9-1a	Parmelia sulcata	92.7	2.71 ± 0.19	2.70 ± 0.19	0.05 ± 0.03	ND	0.996 ± 0.101	0.020 ± 0.012	ND	ND
	L 9-1c	Parmelia acetabulum	81.2	2.77 ± 0.19	3.00 ± 0.20	0.16 ± 0.05	ND	1.08 ± 0.10	0.057 ± 0.018	ND	ND
	L 10-1a	Xanthoria parietina	71.6	1.40 ± 0.14	1.23 ± 0.13	0.07 ± 0.04	ND	0.884 ± 0.130	0.049 ± 0.031	ND	≤ 13.6
	L 10-4a	Xanthoria parietina	58.9	1.79 ± 0.12	1.80 ± 0.12	0.09 ± 0.04	ND	1.00 ± 0.096	0.052 ± 0.020	ND	ND
	F 10-7	—	62.4	1.11 ± 0.12	0.80 ± 0.11	0.03 ± 0.02	ND	0.721 ± 0.124	0.024 ± 0.017	ND	32.1
	F 11-1	—	51.1	0.67 ± 0.10	0.48 ± 0.09	0.02 ± 0.04	ND	0.717 ± 0.175	0.026 ± 0.057	ND	32.6
	L 12-1a	Pseudevernia furfuracea	80.6	1.08 ± 0.10	0.95 ± 0.10	0.05 ± 0.03	ND	0.878 ± 0.123	0.046 ± 0.030	ND	≤ 14.3
	L 12-1c	Parmelia sulcata	101	3.24 ± 0.24	3.47 ± 0.25	0.19 ± 0.07	ND	1.07 ± 0.11	0.057 ± 0.022	ND	ND
	L 12-2a	Physcia biziana	91.2	4.14 ± 0.28	4.69 ± 0.30	0.22 ± 0.07	ND	1.13 ± 0.10	0.052 ± 0.018	ND	ND
	L 12-2b	Parmelia acetabulum	81.9	5.76 ± 0.31	5.32 ± 0.29	0.17 ± 0.06	ND	0.924 ± 0.070	0.029 ± 0.010	ND	≤ 9.1
	L 12-2c	Parmelia borrieri	98.9	4.14 ± 0.33	4.70 ± 0.36	0.33 ± 0.09	ND	1.13 ± 0.12	0.079 ± 0.023	ND	ND

Bratoselce	Ls 18-1	Cladonia convoluta	53.2	82.0±5.2	42.4±2.8	2.23±0.35	0.619±0.156	0.517±0.048	0.027±0.005	0.0075±0.0020	55.3
	Ls 18-1	Cladonia convoluta	73.0	115±3	60.1±1.8	3.31±0.18	0.374±0.047	0.521±0.022	0.029±0.002	0.0032±0.0004	54.8
	L 19-1a	Physcia adscendens	78.5	2.82±0.25	2.82±0.25	0.20±0.07	ND	1.00±0.12	0.070±0.026	ND	ND
	L 20-1a	Physcia adscendens	51.1	2.80±0.28	1.81±0.23	0.13±0.09	ND	0.647±0.103	0.046±0.031	ND	40.5
	L 21-2a	Physcia biziana	80.5	3.08±0.17	2.69±0.15	0.09±0.03	0.023±0.013	0.871±0.068	0.036±0.010	0.0074±0.0042	15.1
	B 21-2a	—	34.0	2.43±0.19	2.38±0.19	0.10±0.06	ND	0.977±0.108	0.041±0.024	ND	ND
	L 21-3a	Parmelia acetabulum	68.4	4.04±0.23	4.25±0.23	0.21±0.06	ND	1.05±0.08	0.053±0.014	ND	ND
	L 21-3a	Parmelia acetabulum	71.8	5.05±0.21	4.75±0.20	0.27±0.04	ND	0.940±0.057	0.054±0.009	ND	≤7.3
	B 21-3a	—	62.1	3.11±0.18	3.10±0.18	0.12±0.04	ND	0.997±0.084	0.040±0.014	ND	ND
	L 21-4	Parmelia borrieri	76.2	3.51±0.20	3.11±0.19	0.09±0.04	0.008±0.010	0.885±0.073	0.025±0.011	0.0023±0.0028	≤13.5
	L 21-4	Parmelia borrieri	98.3	3.89±0.16	4.01±0.16	0.19±0.03	ND	1.03±0.059	0.050±0.008	ND	ND
	B 21-4	—	67.0	3.40±0.19	3.30±0.19	0.20±0.05	ND	0.970±0.079	0.060±0.016	ND	ND
	L 22-1a	Parmelia quercina	95.0	3.01±0.16	2.96±0.16	0.10±0.03	ND	0.985±0.074	0.034±0.011	ND	ND
	L 22-1b	Physcia biziana	92.1	4.36±0.22	4.30±0.22	0.31±0.06	ND	0.985±0.070	0.071±0.014	ND	ND
	L 25-2a	Physcia tenella	78.0	4.89±0.27	5.70±0.30	0.19±0.06	ND	1.16±0.09	0.039±0.012	ND	ND
	L 25-2b	Physconia grisea	66.7	3.50±0.28	3.25±0.27	0.25±0.09	ND	0.927±0.106	0.072±0.025	ND	≤8.7
	L 25-4	—	83.9	0.87±0.07	0.54±0.06	0.06±0.02	0.009±0.006	0.616±0.084	0.065±0.019	0.0098±0.0066	44.0
	L 26-1a	Physconia grisea	67.2	5.83±0.29	5.88±0.29	0.30±0.07	ND	1.01±0.07	0.051±0.012	ND	ND
	L 26-1a	Physconia grisea	94.5	8.50±0.29	8.33±0.28	0.35±0.04	ND	0.980±0.047	0.041±0.005	ND	ND
	B 26-1a	—	66.1	2.37±0.14	2.55±0.14	0.18±0.04	ND	1.08±0.086	0.078±0.018	ND	ND
Reljan	L 28-1a	Physcia adscendens	81.5	2.79±0.17	2.49±0.16	0.11±0.04	0.014±0.011	0.894±0.079	0.044±0.013	0.0049±0.0039	≤12.5
	L 29-1a	Physcia adscendens	59.0	2.65±0.18	2.41±0.17	0.12±0.05	ND	0.908±0.087	0.047±0.018	ND	≤10.9
	B 29-1a	—	40.9	1.02±0.11	1.15±0.12	0.05±0.02	ND	1.13±0.17	0.048±0.024	ND	ND
	Ls 30-1a	Parmelia somloensis	80.1	14.1±0.9	15.2±0.9	0.65±0.14	ND	1.08±0.09	0.046±0.010	ND	ND
	Ls 30-1b	Cladonia convoluta	86.9	15.9±1.1	15.8±1.1	0.99±0.25	ND	0.997±0.100	0.062±0.017	ND	ND
	Lr 31-1	Ramalina capitata	80.0	1.69±0.16	1.62±0.15	0.11±0.05	ND	0.959±0.128	0.063±0.027	ND	≤5.1
	Lr 31-1a	Ramalina capitata	62.1	1.63±0.15	1.82±0.15	0.12±0.04	ND	1.12±0.138	0.071±0.028	ND	ND
	Lr 31-1a	Ramalina capitata	72.1	2.84±0.17	1.68±0.13	0.09±0.03	ND	0.592±0.057	0.030±0.012	ND	46.8
	Lr 31-1a	Ramalina capitata	94.1	2.71±0.14	1.91±0.11	0.10±0.03	ND	0.706±0.055	0.037±0.010	ND	33.8
	Lr 31-1a	Ramalina capitata	97.0	4.30±0.42	2.95±0.36	0.21±0.10	ND	0.685±0.108	0.050±0.023	ND	36.2
M 31-2	—	85.4	10.4±0.34	10.6±0.34	0.49±0.05	ND	1.01±0.04	0.047±0.005	ND	ND	
Mean or range			75.0±16.2					0.949±0.159	0.051±0.019		
				0.22–115	0.28–60.2	0.018–3.31	ND-0.619	0.517–1.32	0.020–0.141	ND-0.0098	ND-55.3

*: not detectable.

Table 8
 ^{238}U , ^{234}U , ^{235}U and ^{236}U concentrations (in Bq kg^{-1}) in biological samples (L: lichen; B: bark; Le: leaf) collected in Montenegro from 27 October to 5 November 2001

Site	Sample code	Lichen species	U yield (%)	^{238}U	^{234}U	^{235}U	^{236}U	$^{234}\text{U}/^{238}\text{U}$	$^{235}\text{U}/^{238}\text{U}$	$^{236}\text{U}/^{238}\text{U}$	F_{DU} , %
Cape Arza	L m1-1a	Xanthoria parietina	108.9	0.89 ± 0.12	0.97 ± 0.12	0.09 ± 0.04	ND	1.08 ± 0.20	0.097 ± 0.050	ND	ND
	LB m1-1	—	73.0	15.2 ± 0.5	7.35 ± 0.30	0.34 ± 0.06	0.07 ± 0.03	0.484 ± 0.026	0.023 ± 0.004	0.0049 ± 0.0018	59.1
	L m2-1a	Parmelia subrudecta	94.5	7.38 ± 0.41	3.59 ± 0.27	0.21 ± 0.07	0.04 ± 0.04	0.486 ± 0.045	0.028 ± 0.010	0.0050 ± 0.0048	58.8
	LB m2-1	—	73.0	11.5 ± 0.4	5.21 ± 0.25	0.34 ± 0.06	0.05 ± 0.03	0.452 ± 0.027	0.030 ± 0.005	0.0043 ± 0.0022	62.8
	Le m2-2	—	71.7	0.25 ± 0.03	0.21 ± 0.03	0.01 ± 0.01	ND	0.850 ± 0.170	0.047 ± 0.050	ND	≤ 17.5
	L m3-1a	Parmotrema chinense	83.2	5.02 ± 0.34	3.53 ± 0.28	0.13 ± 0.06	ND	0.703 ± 0.073	0.026 ± 0.012	ND	34.1
	LB m3-1	—	73.0	8.06 ± 0.33	5.17 ± 0.25	0.39 ± 0.07	ND	0.641 ± 0.041	0.048 ± 0.009	ND	41.2
	Le m3-2	—	80.5	0.30 ± 0.03	0.22 ± 0.03	0.01 ± 0.01	ND	0.724 ± 0.127	0.039 ± 0.037	ND	31.7
	M m4-1	—	102.6	16.4 ± 0.7	10.3 ± 0.5	0.47 ± 0.08	0.11 ± 0.03	0.629 ± 0.038	0.028 ± 0.005	0.0066 ± 0.0021	42.6
	Le m5-1	—	60.6	0.23 ± 0.05	0.16 ± 0.05	0.01 ± 0.02	ND	0.677 ± 0.266	0.053 ± 0.066	ND	37.2
	L m5-2a	Parmotrema chinense	85.1	7.39 ± 0.55	6.68 ± 0.51	0.33 ± 0.11	ND	0.904 ± 0.096	0.044 ± 0.015	ND	≤ 11.4
	L m 5-2b	Parmotrema hypoleucinum	76.8	5.66 ± 0.47	4.58 ± 0.41	0.15 ± 0.08	ND	0.810 ± 0.100	0.027 ± 0.014	ND	22.0
	L m 5-2b	Parmotrema hypoleucinum	71.2	5.03 ± 0.22	5.13 ± 0.22	0.29 ± 0.05	ND	1.02 ± 0.06	0.057 ± 0.009	ND	ND
	LB m5-2	—	73.0	7.60 ± 0.26	5.84 ± 0.22	0.28 ± 0.04	0.03 ± 0.01	0.768 ± 0.039	0.037 ± 0.006	0.0040 ± 0.0019	26.7
	L m6-1a	Parmelia subrudecta	73.0	23.3 ± 0.7	12.0 ± 0.4	0.70 ± 0.06	0.11 ± 0.02	0.515 ± 0.022	0.030 ± 0.003	0.0049 ± 0.0010	55.6
	L m6-1a	Parmelia subrudecta	100.3	14.3 ± 0.5	6.80 ± 0.27	0.32 ± 0.05	0.05 ± 0.02	0.475 ± 0.025	0.022 ± 0.004	0.0035 ± 0.0015	60.1
	L m6-2a	Xanthoria parietina	98.0	15.9 ± 0.5	7.39 ± 0.30	0.52 ± 0.07	0.04 ± 0.02	0.464 ± 0.025	0.033 ± 0.005	0.0025 ± 0.0014	61.3
	B m6-2a	—	58.5	3.65 ± 0.19	1.11 ± 0.10	0.06 ± 0.03	0.01 ± 0.02	0.306 ± 0.031	0.015 ± 0.009	0.0031 ± 0.0056	79.5
	Le m6-4	—	62.5	0.26 ± 0.03	0.20 ± 0.03	0.01 ± 0.01	ND	0.759 ± 0.157	0.055 ± 0.053	ND	27.8
	L m7-1a	Parmelia borrieri	87.0	78.3 ± 4.1	13.2 ± 1.0	0.58 ± 0.19	0.61 ± 0.17	0.169 ± 0.015	0.007 ± 0.002	0.0079 ± 0.0022	95.1
	L m7-1b	Hypogymnia tubulosa	96.8	60.8 ± 2.1	9.85 ± 0.51	0.70 ± 0.13	0.34 ± 0.08	0.162 ± 0.010	0.011 ± 0.002	0.0056 ± 0.0014	95.9
	L m7-1c	Parmotrema chinense	71.5	79.2 ± 4.5	13.1 ± 1.1	1.53 ± 0.34	0.52 ± 0.18	0.165 ± 0.017	0.019 ± 0.004	0.0065 ± 0.0023	95.5
	LB m7-1	—	73.0	73.6 ± 1.9	13.0 ± 0.4	1.15 ± 0.09	0.46 ± 0.05	0.176 ± 0.007	0.016 ± 0.001	0.0062 ± 0.0007	94.3
	Le m7-2	—	74.9	0.25 ± 0.05	0.04 ± 0.04	0.01 ± 0.01	ND	0.162 ± 0.162	0.045 ± 0.049	ND	95.9
	Le m7-3	—	83.4	0.19 ± 0.05	0.13 ± 0.05	0.04 ± 0.02	ND	0.686 ± 0.336	0.204 ± 0.113	ND	36.1
	L m8-1	Parmelia soledians	29.2	36.7 ± 1.5	7.18 ± 0.39	0.58 ± 0.10	0.22 ± 0.06	0.196 ± 0.013	0.016 ± 0.003	0.0059 ± 0.0015	92.1
	L m8-1	Parmelia soledians	61.2	61.2 ± 2.7	11.9 ± 0.7	0.72 ± 0.16	0.42 ± 0.11	0.194 ± 0.015	0.012 ± 0.003	0.0068 ± 0.0018	92.2
	B m8-1	—	101.0	19.6 ± 0.8	3.34 ± 0.25	0.32 ± 0.08	0.11 ± 0.04	0.170 ± 0.015	0.016 ± 0.004	0.0057 ± 0.0022	95.0
	Le m8-2a	—	67.7	0.26 ± 0.05	0.09 ± 0.05	0.02 ± 0.01	ND	0.354 ± 0.191	0.060 ± 0.057	ND	73.9
	Le m8-2b	—	73.0	3.75 ± 0.15	0.88 ± 0.07	0.06 ± 0.02	0.02 ± 0.01	0.234 ± 0.021	0.015 ± 0.005	0.0041 ± 0.0029	87.7
	L m9-1a	Parmotrema chinense	97.1	52.1 ± 1.8	11.1 ± 0.6	0.91 ± 0.16	0.32 ± 0.09	0.214 ± 0.013	0.017 ± 0.003	0.0062 ± 0.0017	90.0
	B m9-1a	—	66.9	11.0 ± 0.6	1.88 ± 0.19	0.14 ± 0.06	0.05 ± 0.03	0.170 ± 0.019	0.013 ± 0.005	0.0043 ± 0.0030	94.9
	LB m9-1	—	73.0	45.8 ± 1.4	11.7 ± 0.5	0.78 ± 0.12	0.24 ± 0.06	0.256 ± 0.014	0.017 ± 0.003	0.0053 ± 0.0014	85.1
	Ls m10-1a	Cladonia rangiformis	95.2	69.4 ± 2.3	10.4 ± 0.4	0.95 ± 0.10	0.36 ± 0.05	0.149 ± 0.008	0.014 ± 0.001	0.0053 ± 0.0008	97.3
	LB m10-1	—	73.0	656 ± 22	85.7 ± 3.2	9.93 ± 0.66	4.90 ± 0.39	0.131 ± 0.007	0.015 ± 0.001	0.0075 ± 0.0006	99.5
	LB m10-1	—	66.8	704 ± 43	93.9 ± 6.1	12.2 ± 1.2	ND	0.133 ± 0.012	0.017 ± 0.002	ND	99.2
L m11-1	—	73.0	45.8 ± 1.3	10.2 ± 0.3	0.90 ± 0.08	0.27 ± 0.04	0.222 ± 0.010	0.020 ± 0.002	0.0059 ± 0.0008	89.1	
L m11-1	—	81.7	22.7 ± 0.9	4.72 ± 0.26	0.51 ± 0.08	0.11 ± 0.03	0.208 ± 0.014	0.022 ± 0.004	0.0050 ± 0.0015	90.6	
Mean or Range			78.0 ± 15.4					0.445 ± 0.285			
				0.19–704	0.04–93.9	0.011–12.2	ND-4.90	0.131–1.08	0.007–0.204	ND-0.0079	ND-99.5

Table 9
 ^{238}U , ^{234}U and ^{235}U concentrations (in $\mu\text{Bq m}^{-3}$) in air filter samples collected in Serbia and Montenegro in 2001

Sample code	Air volume (m^3)	U yield (%)	^{238}U	^{234}U	^{235}U	$^{234}\text{U}/^{238}\text{U}$	$^{235}\text{U}/^{238}\text{U}$	F_{DU} , %
AF 1-1	156	87.4	22.9 ± 1.5	6.20 ± 0.77	0.47 ± 0.27	0.271 ± 0.038	0.021 ± 0.012	83.4
AF 1-3	156	89.6	19.1 ± 1.1	6.77 ± 0.64	0.26 ± 0.18	0.354 ± 0.039	0.014 ± 0.010	74.0
AF 2-1	150	72.8	2.96 ± 0.49	3.26 ± 0.50	0.19 ± 0.19	1.10 ± 0.025	0.064 ± 0.067	ND
AF 2-2	150	74.6	2.33 ± 0.38	2.22 ± 0.38	0.15 ± 0.16	0.953 ± 0.225	0.062 ± 0.068	≤ 5.7
AF 2-3	150	74.5	3.01 ± 0.43	2.01 ± 0.38	0.05 ± 0.15	0.669 ± 0.159	0.018 ± 0.050	38.0
AF 3-1	116	74.2	7.99 ± 0.82	7.87 ± 0.82	0.32 ± 0.26	0.985 ± 0.144	0.040 ± 0.032	ND
AF 3-2	116	56.4	42.1 ± 3.4	38.0 ± 3.2	1.83 ± 0.84	0.901 ± 0.106	0.044 ± 0.020	≤ 11.7
AF 3-3	116	58.0	7.00 ± 0.95	6.82 ± 0.94	0.42 ± 0.36	0.973 ± 0.188	0.060 ± 0.052	ND
AF 4-1	130	56.9	8.37 ± 0.80	7.21 ± 0.74	0.27 ± 0.25	0.861 ± 0.136	0.032 ± 0.030	≤ 16.2
AF 4-2	130	52.5	4.94 ± 0.78	5.44 ± 0.81	0.55 ± 0.36	1.10 ± 0.024	0.112 ± 0.075	ND
AF 4-3	130	62.7	18.8 ± 2.0	19.9 ± 2.1	1.47 ± 0.67	1.06 ± 0.016	0.078 ± 0.037	ND
AF 5-1	130	51.1	10.8 ± 1.1	10.0 ± 1.0	0.33 ± 0.32	0.923 ± 0.133	0.031 ± 0.030	≤ 9.2
AF 5-2	130	47.0	8.68 ± 0.86	8.11 ± 0.83	0.39 ± 0.30	0.934 ± 0.160	0.045 ± 0.035	≤ 7.9
AF 5-3	130	57.7	4.24 ± 0.67	4.59 ± 0.69	0.09 ± 0.24	1.08 ± 0.024	0.020 ± 0.058	ND
AF 6-1	192	63.3	3.04 ± 0.44	1.31 ± 0.34	0.22 ± 0.18	0.429 ± 0.127	0.073 ± 0.060	65.3
AF 6-2	192	67.0	1.99 ± 0.32	0.96 ± 0.26	0.19 ± 0.19	0.483 ± 0.182	0.096 ± 0.098	59.2
AF 6-3	192	63.7	5.02 ± 0.47	1.15 ± 0.28	0.28 ± 0.16	0.229 ± 0.080	0.056 ± 0.041	88.3
Mean or range	116–192	65.1 ± 12.3	1.99–42.1	0.96–38.0	0.05–1.83	0.229–1.10	0.014–0.112	ND–88.3

Table 10
 ^{238}U , ^{234}U and ^{235}U concentrations (in mBq l^{-1}) in tap water (TW), well water (WW) and sea water (SW) samples collected in Serbia and Montenegro in 2001

Sample code	Water volume (l)	U yield (%)	^{238}U	^{234}U	^{235}U	$^{234}\text{U}/^{238}\text{U}$	$^{235}\text{U}/^{238}\text{U}$	F_{DU} (%)
TW6	0.500	86.2	4.54 ± 0.37	5.09 ± 0.39	0.34 ± 0.10	1.12 ± 0.13	0.076 ± 0.023	ND
WW7	0.500	85.6	1.17 ± 0.23	1.31 ± 0.24	0.09 ± 0.06	1.12 ± 0.30	0.077 ± 0.056	ND
TW8	0.500	91.3	16.2 ± 0.8	23.3 ± 1.0	0.83 ± 0.17	1.44 ± 0.10	0.051 ± 0.011	ND
TW13	0.990	61.8	5.20 ± 0.38	5.79 ± 0.40	0.28 ± 0.09	1.11 ± 0.11	0.054 ± 0.018	ND
WW14	1.000	60.9	0.58 ± 0.08	0.74 ± 0.08	0.04 ± 0.02	1.26 ± 0.22	0.074 ± 0.034	ND
WW15	1.000	63.0	0.40 ± 0.10	0.27 ± 0.09	0.01 ± 0.03	0.660 ± 0.277	0.036 ± 0.065	47.7
WW16	1.000	49.5	0.52 ± 0.11	0.43 ± 0.11	0.02 ± 0.03	0.816 ± 0.278	0.036 ± 0.065	32.3
WW17	1.000	54.5	0.61 ± 0.14	0.86 ± 0.15	0.03 ± 0.04	1.41 ± 0.39	0.045 ± 0.064	ND
WW23	1.000	61.3	21.9 ± 0.9	28.1 ± 1.2	0.88 ± 0.14	1.28 ± 0.08	0.040 ± 0.007	ND
WW27	1.000	40.1	0.56 ± 0.12	0.83 ± 0.14	0.02 ± 0.02	1.47 ± 0.40	0.044 ± 0.032	ND
SW12	0.500	75.3	44.4 ± 1.8	49.7 ± 2.0	2.20 ± 0.29	1.12 ± 0.06	0.050 ± 0.007	ND
Mean or range	0.500–1.000	66.3 ± 16.3	0.40–44.4	0.27–49.7	0.01–2.22	0.660–1.47	0.036–0.077	ND–47.7

available bio-indicators to monitor air pollution from potentially toxic substances. In particular, lichens and mosses are efficient accumulators of many elements, including heavy metals and radionuclides that are released into the atmosphere due to natural and human activities. Since the 1960s, they have been successfully employed to assess the global deposition of radionuclides derived from atmospheric nuclear weapons tests and atmospheric re-entry of nuclear powered satellites (Holm and Persson, 1978). A large amount of research has been carried out in particular to monitor

the radionuclides derived from the Chernobyl accident of 1986 (Feige et al., 1990).

The remarkable capacity of lichens for accumulating uranium and other pollutants is based on their intrinsic biology, as they lack roots and outer protective organs, such as stomata and cuticle, do not have access to soil nutrient pools and mainly depend on both wet and dry atmospheric deposition to obtain mineral nutrients. Lichens are usually very long-lived, their morphology does not vary with the seasons and accumulation of pollutants can occur throughout the years. Mosses are

simple green land plants with leaves and a stem and always without roots. Mosses depend upon external moisture to transport nutrients. Many mosses assimilate much of their nutrients and water through their tiny leaves. Unlike the leaves of higher (flowering) plants, which have a waxy covering called a cuticle, mosses lack this protection. This allows mosses to take up water quickly through their leaves during periods of rain or high humidity, and to dry fast when the air dries. This rapid and direct absorption is detrimental when the moisture is laden with pollutants. Study of the elemental composition has provided accord between atmospheric element levels, element concentrations observed in the lichen and moss tissues, and the exposure time of the lichens and mosses to airborne pollutants (UNEP, 2002).

As shown in Table 4, the activity concentrations of uranium isotopes in all tree trunk lichen samples obtained at the control sites in central Italy were detectable, values being in the range of 1.01–4.68 Bq kg⁻¹ for ²³⁸U, 0.85–5.17 Bq kg⁻¹ for ²³⁴U and 0.04–0.28 Bq kg⁻¹ for ²³⁵U with a typical mean activity ratio of 0.984 ± 0.082 for ²³⁴U/²³⁸U and 0.053 ± 0.021 for ²³⁵U/²³⁸U.

The corresponding activity concentrations of uranium isotopes (Table 4) in tree trunk moss samples obtained at the control sites were in the range of 1.18–7.14 Bq kg⁻¹ for ²³⁸U, 1.15–7.69 Bq kg⁻¹ for ²³⁴U and 0.06–0.31 Bq kg⁻¹ for ²³⁵U with a typical mean activity ratio of 1.027 ± 0.082 for ²³⁴U/²³⁸U and 0.057 ± 0.024 for ²³⁵U/²³⁸U. As expected: (1) except for ²³⁸U, ²³⁴U and ²³⁵U, other uranium isotopes in these samples were not detectable and; (2) the uranium isotopic concentrations in moss samples were similar in value to that in lichens due to the fact that mosses are ecologically and structurally closer to lichens than other members of the plant kingdom.

If the ²³⁴U concentrations in lichens and mosses are plotted against the ²³⁸U concentrations (Fig. 2) the correlation equation is $[^{234}\text{U}] = 0.994[^{238}\text{U}] + 0.049$, $r = 0.987$, $n = 30$, $P < 0.001$; the very good linearity correlation shows that the uranium sources in these samples are the same. Fig. 3 shows the correlation between ²³⁵U and ²³⁸U concentrations in these samples, and the obtained equation is $[^{235}\text{U}] = 0.0392[^{238}\text{U}] + 0.0481$, $r = 0.748$, $n = 30$, $P < 0.001$. The results for ²³⁵U are quite scattered due to its lower concentrations and associated with this the high counting uncertainty, but still show a positive correlation.

The radionuclides activity ratios in lichens and mosses vary with the contamination source and they can be utilized to identify the different release sources. The natural composition is characterized by ²³⁴U/²³⁸U and ²³⁵U/²³⁸U activity ratios of about 1.00 and 0.046, respectively. Enriched uranium has higher ²³⁴U/²³⁸U and ²³⁵U/²³⁸U ratios, whereas DU has lower ²³⁴U/²³⁸U

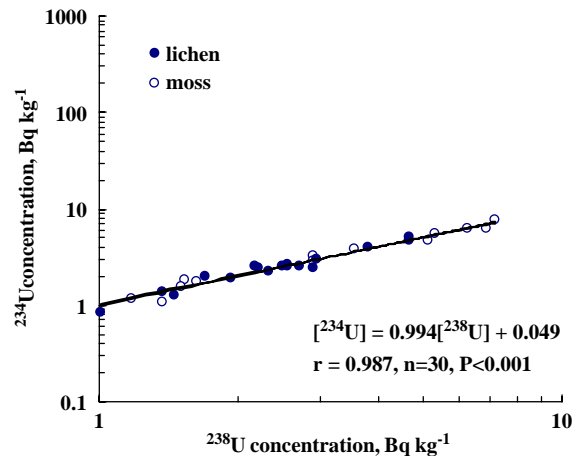


Fig. 2. ²³⁴U concentrations in lichen (●) and moss (○) samples collected from Urbino (Central Italy) as a function of their ²³⁸U concentration.

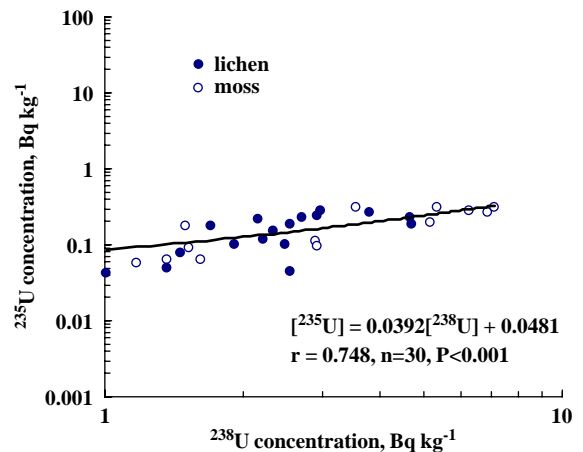


Fig. 3. ²³⁵U concentrations in lichen (●) and moss (○) samples collected from Urbino (Central Italy) as a function of their ²³⁸U concentration.

and ²³⁵U/²³⁸U ratios. The average ratios of ²³⁴U/²³⁸U and ²³⁵U/²³⁸U in these lichens and mosses obtained from Table 4 are 1.009 ± 0.098 and 0.058 ± 0.024, respectively. The value of the ratio ²³⁴U/²³⁸U of close to unity shows that the uranium in the samples is mainly natural, but there is a possible uranium contribution from fallout of atmospheric nuclear weapon tests conducted in the 1950s and 1960s due to the higher ²³⁵U/²³⁸U ratio (>0.046).

Due to their complex surface, slow growth rate and slow metabolic activity, lichen and mosses are known to strongly accumulate a great variety of radionuclides

from atmospheric contamination with a very slow loss. Early studies showed that the effective half-life of ^{137}Cs was 10 y in lichens and 4 y in mosses (Mihok et al., 1989); as such, it seems that lichens are more effective than mosses as accumulators of ^{137}Cs airborne pollution. Conversely, from the data shown in Figs. 2 and 3 it is seen that the uranium concentrations in these lichen and moss samples are of the same level, therefore, the accumulation effects of uranium in tree trunk lichens and mosses are not distinguishable.

The uranium isotopic concentrations in tree trunk lichen and moss, mushroom and bark samples collected in Serbia and Montenegro are shown in Table 7 and 8, values being in the range of $0.67\text{--}704\text{ Bq kg}^{-1}$ for ^{238}U , $0.48\text{--}93.9\text{ Bq kg}^{-1}$ for ^{234}U and $0.02\text{--}12.2\text{ Bq kg}^{-1}$ for ^{235}U , showing that the uranium levels are higher than in the samples collected at the control sites (Table 4). Moreover, ^{236}U is detectable in some of the samples. The activity concentration ratios are in the range of $0.131\text{--}1.32$ for $^{234}\text{U}/^{238}\text{U}$, $0.007\text{--}0.141$ for $^{235}\text{U}/^{238}\text{U}$, and not detectable (ND) -0.0098 for $^{236}\text{U}/^{238}\text{U}$, showing the existence of DU in some samples. Due to the contribution from the substrate, the uranium activity concentrations in lichens collected from soil or rock are generally higher than in the tree trunk lichens, which are in the range of $1.63\text{--}20.3\text{ Bq kg}^{-1}$ for ^{238}U , $1.62\text{--}21.3\text{ Bq kg}^{-1}$ for ^{234}U and $0.10\text{--}1.30\text{ Bq kg}^{-1}$ for ^{235}U ; the activity concentration ratios are in the range of $0.592\text{--}1.12$ for $^{234}\text{U}/^{238}\text{U}$ and $0.030\text{--}0.077$ for $^{235}\text{U}/^{238}\text{U}$. These results also indicate the presence of DU in some of the samples. The highest uranium activity concentrations were found in the samples collected at the Cape Arza site in Montenegro (704 Bq kg^{-1} for ^{238}U). It is noted that at this site not only are the uranium contents higher, but also DU is detectable in nearly all the samples. These results reflect the intense battle that occurred during the conflict at this site, the DU penetrators hitting hard surfaces, shattering into dust and then being dispersed in the air.

Pine tree leaves were also collected at the Cape Arza site. The uranium isotopic concentration in the leaf samples were much lower than that in other biological samples, and were in the range of $0.19\text{--}3.75\text{ Bq kg}^{-1}$ for ^{238}U , $0.04\text{--}0.88\text{ Bq kg}^{-1}$ for ^{234}U and $0.01\text{--}0.06\text{ Bq kg}^{-1}$ for ^{235}U . The activity concentration ratios were in the range of $0.162\text{--}0.850$ for $^{234}\text{U}/^{238}\text{U}$ and $0.015\text{--}0.204$ for $^{235}\text{U}/^{238}\text{U}$. The samples were also all DU positive.

Based on Eq. (2), the DU fractions in all the biological samples have been calculated and the relevant results are also given in Tables 7 and 8. It is shown that DU is detectable at all seven monitoring sites, the highest DU fraction of 99.5% being found in a mixed lichen and bark sample at the Cape Arza site.

The mechanisms of trace element uptake and retention in lichens differ from species to species. In order to

distinguish the accumulation effect of uranium among lichen species, the lichens have been classified as shown in Tables 7 and 8. However, due to: (1) the limited sample numbers, and; (2) large variation of the uranium concentrations even in the same species, statistical evaluation of the uranium accumulation effects in different lichen species are still not possible. The information is nevertheless very helpful for future studies on the mechanism and accumulation effects of DU in different lichen species.

Figs. 4 and 5 show the correlation between ^{234}U or ^{235}U and ^{238}U activity concentration in all the biological samples collected in Serbia and Montenegro, being either expressed as a power regression as $[^{234}\text{U}] = 0.941[^{238}\text{U}]^{0.777}$, $r = 0.949$, $n = 108$, $P < 0.001$ and $[^{235}\text{U}] = 0.0527[^{238}\text{U}]^{0.794}$, $r = 0.942$, $n = 108$, $P < 0.001$ respectively, or by linear regression as $[^{234}\text{U}] = 0.134[^{238}\text{U}] + 3.90$, $r = 0.906$, $n = 108$, $P < 0.001$ and $[^{235}\text{U}] = 0.0162[^{238}\text{U}] + 0.132$, $r = 0.985$, $n = 108$, $P < 0.001$. Comparing the linear correlation between ^{234}U and ^{238}U concentrations in Fig. 4 with that in Fig. 2, less good correlation is obtained in Fig. 4. The high variability found in the activity ratios of $^{234}\text{U}/^{238}\text{U}$ is as a result of DU contamination in the samples, indicating that the radiological impact of DU ordnance was site-specific and that the contamination scenario was different in the various parts of the investigated area. Carefully examining the results in Fig. 4, it seems that the results can be classified into two or three parts. The results in the upper part can be expressed by a linear regression as $[^{234}\text{U}] = 1.04[^{238}\text{U}] - 0.0947$, $r = 0.997$, $n = 55$, $P < 0.001$, which represent the samples uncontaminated by DU or containing mainly natural uranium similar to Fig. 2; the results in the middle and lower parts concern samples contaminated from DU or containing both

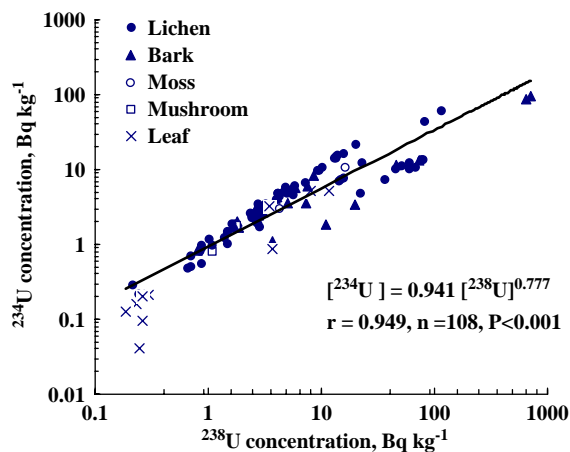


Fig. 4. ^{234}U concentrations in lichen (●), bark (▲), moss (○), mushroom (□) and leaf (×) samples collected from Serbia and Montenegro as a function of their ^{238}U concentrations.

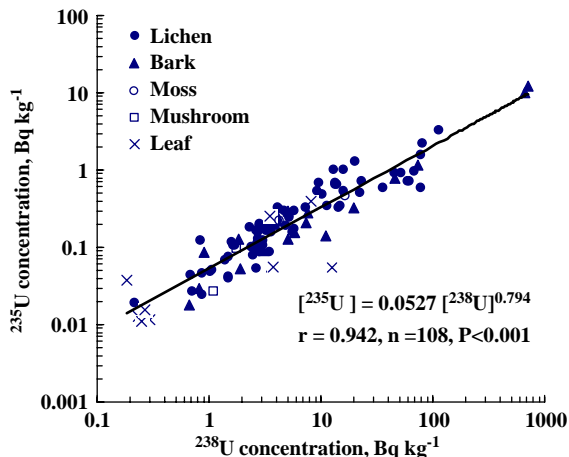


Fig. 5. ^{235}U concentrations in lichen (●), bark (▲), moss (○), mushroom (□) and leaf (×) samples collected from Serbia and Montenegro as a function of their ^{238}U concentrations.

natural and DU. In summary, Figs. 4 and 5 provide a representative description of uranium isotopic ratios for the current environmental ecosystem in Serbia and Montenegro following the Balkan conflict in 1999. Moreover, the results from the field survey in Serbia and Montenegro have clearly demonstrated that lichens, mosses, mushrooms, leaves and barks are sensitive bio-indicators of past airborne contamination of DU dust or particles, in particular in areas where DU ammunition has been used.

3.2. Uranium in air

The concentrations of uranium in air vary widely, depending on the weather conditions and the uranium content in soil. Example literature values are $0.9\text{--}5\ \mu\text{Bq m}^{-3}$ in the United States and $0.02\text{--}18\ \mu\text{Bq m}^{-3}$ in Europe (UNSCEAR, 2000). The given reference value is $1\ \mu\text{Bq m}^{-3}$ of ^{238}U . Most of the natural uranium levels will be in the range of $0.1\text{--}10\ \mu\text{Bq m}^{-3}$.

As presented in Table 9, the uranium isotopic concentrations in air obtained from the air filter samples collected in Serbia and Montenegro are in the range of $1.99\text{--}42.1\ \mu\text{Bq m}^{-3}$ for ^{238}U , $0.96\text{--}38.0\ \mu\text{Bq m}^{-3}$ for ^{234}U , and $0.05\text{--}1.83\ \mu\text{Bq m}^{-3}$ for ^{235}U , which are in the typical range of natural uranium values.

The highest value (AF 3-2) was found in the samples collected at Bratoselce. At this site the highest value of natural uranium concentrations in soil were also measured (UNEP, 2002). The lowest uranium concentrations and also the lowest $^{234}\text{U}/^{238}\text{U}$ activity ratios were measured in Cape Arza, which is located near the sea. It is well known that at coastal locations uranium concentrations in air may be an order of magnitude

lower than in continental or industrialized areas, which explains the low uranium concentration.

Based on Eq. (2), the DU fractions in air samples have also been calculated (Table 9). It is interesting to note that although the detected uranium isotopic concentrations in all the collected air samples are within the typical range of natural uranium values, the $^{234}\text{U}/^{238}\text{U}$ activity ratio analyses strongly indicate DU to be contained in two air samples from Pljackovica (AF 1-1 and AF 1-3), one each from Borovac (AF 2-3), Bratoselce (AF 3-2) and Bukurevac (AF 4-1), and three from Cape Arza (AF 6-1, AF 6-2, AF 6-3). The specific activities of DU in the samples at the Pljackovica and Cape Arza sites have been found to be higher than the specific activities of natural occurring uranium. The existence of DU in air can only be explained by resuspension of DU contaminated particles and/or soil. This fact agrees well with the widespread DU contamination detected in the biological samples.

The risk assessment to the public from exposure to DU is mainly associated with its radiological (external and internal exposures) and chemical effects (chemical toxicity), with dependency on physical and chemical behaviour of DU, concentration level in the environment, uptake in the body and so on. As far as radiological risk is concerned, inhalation of DU from air is one of the most important pathways. Based on the respiratory volume of air for reference man ($23\ \text{m}^3\ \text{d}^{-1}$), the committed effective dose conversion factors for uranium isotopes (insoluble uranium: slow clearance from lung) ($8.0 \times 10^{-6}\ \text{Sv Bq}^{-1}$ for ^{238}U , $9.6 \times 10^{-6}\ \text{Sv Bq}^{-1}$ for ^{234}U and $8.5 \times 10^{-6}\ \text{Sv Bq}^{-1}$ for ^{235}U) and the DU concentration in air (in $\mu\text{Bq m}^{-3}$), the annual committed effective dose due to the inhalation of DU from air as estimated on a worst-case scenario assuming that a person lives continuously in the place where the maximum concentration of DU in air was detected (Pljackovica) is $1.73\ \mu\text{Sv}$ ($1.28\ \mu\text{Sv}$ from ^{238}U , $0.42\ \mu\text{Sv}$ from ^{234}U and $0.03\ \mu\text{Sv}$ from ^{235}U) (ICRP, 1974; EU, 1996). The ICRP-recommended subsidiary dose limit for critical groups and the principal dose limit for public are 5 and $1\ \text{mSv y}^{-1}$, respectively (ICRP, 1977). Therefore, the dose due to the inhalation of DU from air in Serbia and Montenegro is insignificant.

One also has to consider that the air samples were collected under dry warm weather condition at moderate wind speed. At higher wind speed the concentrations of DU in air might be higher, while in damp, rainy weather the DU particles are washed out from the air and deposited on the ground. However, given the entire range of climatic possibilities, the DU concentrations in air at the investigated sites would almost certainly have never been so high that the resulting dose would have been significant.

3.3. Uranium in water

Uranium concentrations in natural waters vary from region to region due to the different rocks composing the aquifer, the water composition, the distance from uraniumiferous areas and climatic and agricultural conditions that prevail. The values ranged widely, from <0.01 to 150 Bq l^{-1} . It is reported that the typical $^{234}\text{U}/^{238}\text{U}$ activity ratios in natural water samples range from 0.8 to 10, while $^{235}\text{U}/^{238}\text{U}$ activity ratio is thought to have a quite uniform value of about 0.046 (Goldstein et al., 1997). For comparison Table 6 shows the uranium isotopic concentrations in drinking water, filtered river water and seawater samples collected in central Italy as control sites. It is seen that the uranium concentrations in Tireno and Adriatic seawaters are medium high and constant with a mean ratio of 1.15 ± 0.06 for $^{234}\text{U}/^{238}\text{U}$ and 0.054 ± 0.008 for $^{235}\text{U}/^{238}\text{U}$. Conversely, the uranium concentrations in drinking and river waters vary considerably and range from 0.30 to 103 mBq l^{-1} for ^{238}U , from 0.49 to 135 mBq l^{-1} for ^{234}U , and from 0.02 to 4.82 mBq l^{-1} for ^{235}U . The mean activity ratios are 1.35 ± 0.19 for $^{234}\text{U}/^{238}\text{U}$ and 0.050 ± 0.009 for $^{235}\text{U}/^{238}\text{U}$. All the data in Table 6 indicate characteristics of natural uranium. The WHO have derived a guideline for drinking water uranium quality of $2 \mu\text{g l}^{-1}$ ($24.9 \text{ mBq }^{238}\text{U l}^{-1}$) and the value is considered to be protective for sub-clinical renal effects reported in epidemiological study (WHO, 1998). From Table 6 it is seen that the uranium concentrations in some drinking waters are above the derived guideline value. Most of the drinking waters in Table 6 are mineral water. Geological conditions, where there is high radiation background, high concentration of organic matter, iron hydroxide, carbonaceous material, clay minerals or sulphides, are the most important factors for the uranium concentration variation in drinking water. In some parts of Italy, especially in the volcanic regions, minerals contain high level of natural uranium and thorium. Due to the complex and/or redox reactions in water, some uranium can be leached out and take on a soluble form, for instance, uranyl carbonate (UO_2CO_3) which is formed by the action of CO_2 under pressure on UO_2^{2+} and stable up to 500°C (Partington, 1966). This could be the reason why the uranium concentrations in some Italian mineral waters are significant.

During the field mission in Serbia and Montenegro great attention has been paid to the possible water contamination of DU. The results of the uranium assay in the water samples collected in Serbia and Montenegro are presented in Table 10. From these results, it can be concluded that the uranium concentrations are much lower than those in mineral water found in central Italy (Table 6). The activity concentrations range from 0.40 to 21.9 mBq l^{-1} for ^{238}U , from 0.27 to 28.1 mBq l^{-1} for ^{234}U and from 0.01 to 0.88 mBq l^{-1} for ^{235}U . The mean

activity ratios range from 0.660 to 1.47 for $^{234}\text{U}/^{238}\text{U}$ and from 0.036 to 0.077 for $^{235}\text{U}/^{238}\text{U}$. The low concentrations and solubility of uranium in rocks and soils are the cause of low uranium concentration in the waters. The activity ratios for these samples, except for WW15 and WW16, are consistent with a predominantly natural source of uranium for almost the entire set of samples. However, two samples collected from a private well at Borovac (WW15 and WW16) show low $^{234}\text{U}/^{238}\text{U}$ activity ratios of 0.660 and 0.816, and the estimated DU fractions (m/m) in the two samples are 47.7 and 32.3%, respectively, indicating the possible presence of anthropogenic contribution of DU in the samples. The low uranium concentrations associated with the high relative uncertainty in the two samples reflects poor counting statistics, therefore further investigation on the two sites is necessary.

Based on the information currently available, the uranium concentrations in Serbia and Montenegro waters are below the guideline derived by WHO for drinking water. Therefore, the uranium isotopes in these waters do not constitute a risk to health at the present time from the radiotoxicological point of view. However, in some cases the aquifer depth is very close to the ground surface, as a result of which the filtering action of soil might be reduced and DU leached from the buried penetrators and fragments might reach the groundwater system. Previous study has indicated that DU, when it occurs in small fragments or as dust particles, is more easily dissolved than uranium in mineral lattices (Sansone et al., 2001). Thus, it has higher mobility than natural uranium and the extent of the migration of the dissolved DU through the soil profile could represent a potential risk of future contamination of the underground aquifers. Since only limited numbers of samples have been collected in the areas where DU ordnance was used, groundwater used for drinking should be checked in future to increase the confidence with regard to results observed during the fields studies in Serbia and Montenegro (UNEP, 2002).

4. Conclusion

The uranium isotopic concentrations in biological samples collected in Serbia and Montenegro, mainly lichens and barks, are in the range of 0.67 – 704 Bq kg^{-1} for ^{238}U , 0.48 – 93.9 Bq kg^{-1} for ^{234}U and 0.02 – 12.2 Bq kg^{-1} for ^{235}U , showing that the uranium levels are higher than in the samples collected at the control site. Moreover, the ^{236}U in some of the samples is detectable. The isotopic ratios of $^{234}\text{U}/^{238}\text{U}$ show that DU is detectable in many biological samples at all examined sites, especially in Montenegro, indicating the widespread ground-surface DU contamination, albeit at very low level. The presence of DU in the biological

samples was as a result of DU contamination in air. The air contamination sources could be: (1) direct DU contamination, i.e. at least some of the penetrators hit hard surfaces, shattered into dust and dispersed in air during the conflict, and/or; (2) indirect DU contamination, i.e. resuspension of DU contaminated surface soil after the conflict. The radiological survey also shows that lichens, and in addition barks, are sensitive bio-indicators of past airborne contamination of DU dust or aerosol particles generated at the time of conflict by DU penetrators.

The uranium isotopic concentrations in air obtained from the air filter samples collected in Serbia and Montenegro are in the range of 1.99–42.1 $\mu\text{Bq m}^{-3}$ for ^{238}U , 0.96–38.0 $\mu\text{Bq m}^{-3}$ for ^{234}U and 0.05–1.83 $\mu\text{Bq m}^{-3}$ for ^{235}U , which are in the typical range of natural uranium values. Thus said, most of the air filter samples are DU positive, a fact agreeing well with the widespread DU contamination detected in the biological samples.

The uranium concentrations in water samples collected in Serbia and Montenegro are in the range of 0.40–21.9 mBq l^{-1} for ^{238}U , 0.27–28.1 mBq l^{-1} for ^{234}U and 0.01–0.88 mBq l^{-1} for ^{235}U , which are much lower than those in mineral water found in central Italy and below the guideline derived by WHO for drinking water.

From a radiotoxicological of view, at this moment there is no significant radiological risk related to these investigated sites in terms of possible DU contamination of water, air and/or plants.

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