



## Uranium Isotopes, Metals and Other Elements in Lichens and Tree Barks Collected in Bosnia–Herzegovina

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**Abstract.** The United Nations Environment Programme (UNEP) performed field surveys at 15 sites in Bosnia–Herzegovina where depleted uranium (DU) ammunition was used by the North Atlantic Treaty Organization (NATO) during the Balkans conflict (1994–1995). During the field missions, the Italian Environmental Protection Agency (APAT) evaluated airborne contamination due to DU dusts or aerosol particles, generated at the time of the conflict by the impact of DU ammunition on hard “targets,” using lichens and tree barks as biomonitors. Each sample was analyzed by alpha-spectrometry for DU determination. The <sup>234</sup>U/<sup>238</sup>U activity concentration ratios were used to distinguish natural from anthropogenic uranium. This paper reports the data obtained by the UNEP investigation, including (non-radioactive) metal and other element concentrations in lichen and tree bark samples measured by instrumental neutron activation analysis (INAA). The results indicated: (i) lichens and tree barks are sensitive bio-accumulators of past airborne contamination by depleted uranium dusts; (ii) 8 years after the conflict, environmental DU contamination is still present at some of the target sites; and (iii) the highest concentrations of most non-radioactive elements were found at sites used for ammunition destruction.

**Key words:** depleted uranium, biomonitoring, air pollution, Balkans region.

### 1. Introduction

In July 2000, North Atlantic Treaty Organization (NATO) provided United Nations Environment Programme (UNEP) with information on the use of DU ammunition during the conflict in the Balkan region (1994–1995). On this basis, in October 2000 UNEP started a series of surveys to assess the environmental impact of DU when used in a real conflict situation.

The field mission in Bosnia–Herzegovina (October 2002) represents the final DU assessment of this programme (UNEP, 2001, 2002, 2003).

Natural uranium is composed of three isotopes: 99.275% uranium-238 ( $^{238}\text{U}$ ), 0.72% uranium-235 ( $^{235}\text{U}$ ) and 0.005% uranium-234 ( $^{234}\text{U}$ ). Depleted uranium is a byproduct of the uranium enrichment process applied in the production of nuclear fuel; it has a  $^{235}\text{U}$  content of only 0.2–0.4%. Due to its high density, availability, and relatively low cost, uranium has been incorporated into projectiles and armor.

When a DU projectile hits a hard object, e.g. an armoured vehicle, the penetrator pierces the metal sheet, generally leaving the jacket behind. The DU dust which may be formed during the impact can be dispersed and contaminate the environment. It is estimated that normally 10–35% (and a maximum of 70%) of the DU penetrator becomes an aerosol on impact or when the DU catches fire. Most of the dust particles have been reported to be smaller than  $5\ \mu\text{m}$  in size which keeps them airborne for an extended time, and spread according to wind direction (Bleise *et al.*, 2003).

In the environment, DU has the same mechanism of diffusion as natural uranium, depending on its chemical and physical form (chemical species, particle dimensions, solubility, etc.). Once deposited on the ground, it is subject to dispersion according to wind direction, and to leaching depending on soil characteristics and on the meteorological and climatic conditions of the site (Sansone *et al.*, 2001b).

This paper reports a survey of uranium isotopes, metals and some other elements in biological samples collected in Bosnia–Herzegovina during the field study conducted by UNEP in 2002. Atmospheric deposition was evaluated by collecting lichens and tree barks. Lichens are commonly used to assess and monitor air quality because they are perennial, slow-growing organisms that accumulate elements, including uranium, mainly by trapping atmospheric particulates (Garty, 1992). Tree barks accumulate atmospheric pollutants over long periods of time through wet and dry deposition; the bark surface is very porous, and the absence of a metabolic process makes it almost inert in the presence of inorganic and organic substances (Musilek *et al.*, 2000; Bellis *et al.*, 2001).

## 2. Materials and Methods

### 2.1. INVESTIGATED SITES

UNEP selected 15 sites to be visited during the mission (Figure 1); five sites (1, 4, 5, 6 and 7) where NATO fired DU munitions and the remaining sites where the local population or authorities were concerned that DU might have been used. A detailed description of the investigated sites where botanical samples were collected is reported elsewhere (UNEP, 2003; Rosamila *et al.*, 2003); at sites 5, 8, 9 and 13, for different reasons (mine-field, lichens not present, etc.), biological samples were not collected.

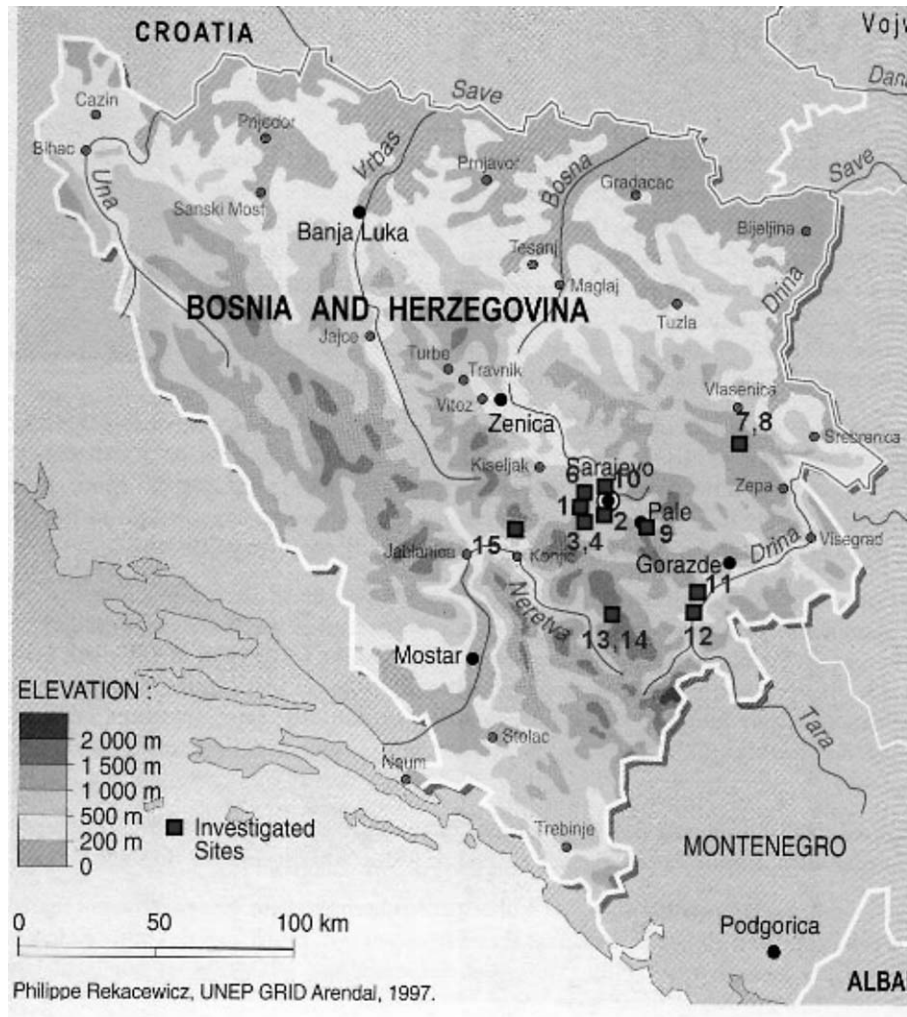


Figure 1. Investigated sites at Bosnia and Herzegovina.

## 2.2. LICHEN AND TREE BARK SAMPLES

Samples of tree bark and/or epiphytic lichens were collected at each site – whenever this was feasible due to the local situation as regards mines – in order to search for the presence of DU dust particles and metals.

According to the “guidelines for the use of epiphytic lichen as biomonitors of atmospheric deposition of trace metals” (Nimis and Bargagli, 1999), lichens were collected by choosing:

- fruticose or foliose broad-lobed species only;
- epiphytic species whenever possible;

- mature trees, with growth of bryophytes not higher than 25%;
- lichens from all around the trunk;
- samples at a height of more than 1 m above the ground, to avoid terrigenous contamination.

At each location, epiphytic lichens were collected from trees and/or bushes using a steel knife and placed in paper envelopes. Samples of tree bark were also collected at some sites to investigate the relationship between the uranium content in bark and lichen samples. After collection, the botanical samples were air-dried and then stored in paper bags (Rosamilia *et al.*, 2003).

The collected lichen species were identified in the laboratory; the nomenclature follows Nimis (2003).

Mainly foliose epiphytic lichen species *Hypogymnia physodes* (10 samples), *Parmelia sulcata* (7), *Hypogymnia tubulosa* (1), *Xantoria parietina* (1), *Platismatia glauca* (1), one fruticose specie (*Evernia prunastri*) and one saxicolous specie (*Squamarina* cfr. *stella-petraea*) were identified among the collected samples.

### 2.3. SAMPLE PREPARATION

Lichen samples were separated from the bark substratum using a steel knife and extraneous material such as mosses and large soil particles was removed under a binocular microscope. The samples were not washed as the aim was to measure the elements that were physically trapped on the surface of the thallus, as well as chemically bound to cell walls (Fahselt, 1997).

Soil, insects or any other solid matter was removed from bark samples. Using a hard steel knife, the external surface (about 3 mm) of barks was cut and dried at 40° C for 24 h. A fine powder ( $\leq 250 \mu\text{m}$ ) of lichen and tree bark samples was obtained by grinding in a Planetary Ball Mill (Retsch PM4), using agate grinding jars of 500 ml with agate balls of 20 mm diameter (Rusu, 2002). Agate is generally used to avoid lead contamination of samples.

Samples were stored in polyethylene containers. A Teflon ball was put into each container when initially filled to facilitate subsequent re-homogenisation.

### 2.4. DU DETERMINATION

$^{238}\text{U}$ ,  $^{234}\text{U}$  and  $^{235}\text{U}$  concentrations in all samples were determined by alpha-spectrometry (CANBERRA) following total dissolution (Sansone *et al.*, 2001a; Jia *et al.*, 2002a, b). For each sample one analysis was performed.

The energy calibration of the alpha spectrometer was performed with  $^{241}\text{Am}$ ,  $^{244}\text{Cm}$  and  $^{237}\text{Np}$ . The efficiency calibration was performed with  $^{232}\text{U}$  or  $^{236}\text{U}$ . A known amount of  $^{232}\text{U}$  tracer ( $^{232}\text{U}$  is not present in the sample) was added to each solution. In evaluating the spectra, the count rate of the tracer can be directly compared with the count rate of the unknown amount of uranium isotopes in the

sample. Any loss of sample will be compensated by this technique. Based on the count rates of the tracer and the unknown, the content in Bq/kg can be calculated.

Usually the analytical procedure to dissolve botanical samples is less aggressive than the procedure used to dissolve soil/sediment sample. As known, lichens and tree barks can accumulate DU by trapping soil particles and different types of air-suspended particles (pollen, dust, etc.). The soil-dissolving procedure was selected for lichen samples in order to compare the values found in lichen with those found in soil samples. The accuracy of the method was assessed using IAEA 326 Soil RM. The repeatability was 3% for  $^{238}\text{U}$  and 1.1% for  $^{234}\text{U}$ .

As in the previous UNEP missions, the team members decided to organise a quality control exercise based on an inter-laboratory comparison (using the Reference Materials IAEA 336 “Lichen” and IAEA 326 “Soil”) between the APAT Laboratory (Italy) and the Spiez Laboratory (Switzerland) in order to verify the precision and comparability of the analytical results obtained in the two laboratories. The repeatability of the Spiez laboratory was 2.3% for  $^{238}\text{U}$ . The results of the quality control exercise between the both laboratories showed good agreement. A detailed description of the sampling locations, biological sampling, sample preparation methods and the results of the quality control exercise are reported in the UNEP 2003 and APAT Reports (Rosamilia *et al.*, 2003).

## 2.5. DETERMINATION OF METALS

From about 100–200 mg of powdered lichen or bark samples tablets were made in a die using a SPECAC Hydraulic Press and then used for instrumental neutron activation analysis (INAA). Multielemental INAA, so-called  $k_0$ -based INAA, was applied for determination of metals and other elements in all samples (Jeran and Jačimovič, 2001). Tablets were irradiated together with Al–0.1% Au discs serving as fluence rate monitors for 20 h in the TRIGA Mark II reactor of the “Jožef Stefan” Institute (JSI), Ljubljana, Slovenia, at a thermal neutron fluence rate of  $1.1 \times 10^{16} \text{ nm}^{-2} \text{ s}^{-1}$ . Gamma spectrometric measurements were performed after 3 and 8 days cooling time on absolutely calibrated high-purity (HP) Ge-detectors. For evaluation of gamma spectra, Hypermet-PC software was used, and for the calculation of elemental concentrations, the KAYZERO/SOLCOI (1996) program was applied. Analytical quality control was applied by analysing the standard reference material IAEA 336 “Lichen”. Accuracy for As, Ba, Ca, Co, Cr, Fe, Sb, Sc, Th, U, and Zn was within 10%.

## 3. Results and Discussion

### 3.1. DEPLETED URANIUM

Table I reports the activity concentration of  $^{238}\text{U}$ ,  $^{234}\text{U}$  and  $^{235}\text{U}$ , expressed on a dry weight basis, and the values of  $^{234}\text{U}/^{238}\text{U}$  activity ratios measured in lichens and tree barks.

Table I. Concentrations of  $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{234}\text{U}/^{238}\text{U}$  ratios in samples collected in Bosnia-Herzegovina

SITE	Sample type	$^{238}\text{U}$ Bq/kg	$^{234}\text{U}$ Bq/kg	$^{235}\text{U}$ Bq/kg	$^{234}\text{U}/^{238}\text{U}$
1	Bark and <i>Phaeophyscia orbicularis</i> (Neck.) <sup>(1)</sup> Moberg	1.83 ± 0.08	1.59 ± 0.07	0.10 ± 0.02	0.87 ± 0.05
	<i>Hypogymnia physodes</i> (L.) Nyl.	1.9 ± 0.1	1.78 ± 0.09	0.08 ± 0.02	0.94 ± 0.07
	<i>Melanelia subaurifera</i> (Nyl.) Essl.	2.6 ± 0.1	2.6 ± 0.1	0.19 ± 0.03	0.97 ± 0.06
	<i>Parmelia sulcata</i> Taylor	2.29 ± 0.09	2.02 ± 0.08	0.12 ± 0.02	0.88 ± 0.05
	<i>P. sulcata</i>	2.17 ± 0.09	1.67 ± 0.08	0.08 ± 0.02	0.77 ± 0.05
	<i>P. sulcata</i>	3.6 ± 0.1	2.4 ± 0.1	0.15 ± 0.02	0.67 ± 0.03
	Bark	0.99 ± 0.04	0.84 ± 0.04	0.04 ± 0.01	0.85 ± 0.05
	Bark and <i>Arthopyrenia</i> sp. <sup>(1)</sup>	1.10 ± 0.05	0.93 ± 0.05	0.05 ± 0.01	0.85 ± 0.06
	Bark and <i>Arthopyrenia</i> sp. <sup>(1)</sup>	1.73 ± 0.07	1.45 ± 0.07	0.11 ± 0.02	0.84 ± 0.05
2	Bark	0.84 ± 0.04	0.67 ± 0.03	0.06 ± 0.01	0.80 ± 0.06
	<i>P. sulcata</i>	2.41 ± 0.09	2.31 ± 0.09	0.15 ± 0.02	0.96 ± 0.05
	<i>Physcia adscendens</i> Fr. H. Oliver	2.9 ± 0.1	2.8 ± 0.1	0.16 ± 0.03	0.94 ± 0.06
3	<i>P. sulcata</i>	1.86 ± 0.08	1.72 ± 0.08	0.10 ± 0.02	0.93 ± 0.06
	<i>H. physodes</i>	1.20 ± 0.07	1.36 ± 0.07	0.07 ± 0.02	1.14 ± 0.09
4	Bark	0.35 ± 0.02	0.24 ± 0.02	0.02 ± 0.01	0.67 ± 0.06
	<i>H. physodes</i>	10.2 ± 0.3	2.5 ± 0.2	0.26 ± 0.06	0.25 ± 0.02
	<i>P. sulcata</i>	29.7 ± 1.1	5.8 ± 0.4	0.7 ± 0.2	0.19 ± 0.01
6	<i>H. physodes</i>	1.20 ± 0.06	1.26 ± 0.06	0.07 ± 0.02	1.05 ± 0.07
	<i>P. sulcata</i>	2.0 ± 0.1	1.9 ± 0.1	0.12 ± 0.03	0.97 ± 0.07
	<i>H. physodes</i>	1.21 ± 0.08	1.31 ± 0.08	0.05 ± 0.02	1.08 ± 0.09
7	<i>P. sulcata</i>	2.71 ± 0.08	1.97 ± 0.07	0.09 ± 0.02	0.73 ± 0.03
	<i>Evernia prunastri</i> (L.) Ach.	1.4 ± 0.1	1.3 ± 0.1	0.05 ± 0.03	0.9 ± 0.1
	<i>H. physodes</i>	1.54 ± 0.08	1.11 ± 0.06	0.07 ± 0.02	0.72 ± 0.05
	<i>Platismatia glauca</i> (L.) W.L. Culb. and C.F. Culb.	1.23 ± 0.05	1.05 ± 0.04	0.07 ± 0.01	0.85 ± 0.05
	<i>H. physodes</i>	1.30 ± 0.06	0.74 ± 0.04	0.08 ± 0.02	0.57 ± 0.04
	<i>H. physodes</i>	1.36 ± 0.06	0.94 ± 0.05	0.07 ± 0.02	0.69 ± 0.05
10	<i>Parmelia saxatilis</i> (L.) Ach.	2.36 ± 0.09	1.29 ± 0.06	0.05 ± 0.02	0.55 ± 0.03
	Bark (Schaer.) Hav.	1.22 ± 0.05	1.24 ± 0.05	0.03 ± 0.01	1.02 ± 0.06
	<i>Hypogymnia tubulosa</i>	1.07 ± 0.07	1.06 ± 0.07	0.06 ± 0.02	0.99 ± 0.09
	<i>H. physodes</i>	0.93 ± 0.06	0.94 ± 0.06	0.02 ± 0.01	1.01 ± 0.09

(Continued on next page.)

Table I. (Continued)

SITE	Sample type	$^{238}\text{U}$ Bq/kg	$^{234}\text{U}$ Bq/kg	$^{235}\text{U}$ Bq/kg	$^{234}\text{U}/^{238}\text{U}$
11	<i>Xanthoria parietina</i> (L.) Th.Fr.	$2.58 \pm 0.09$	$2.48 \pm 0.09$	$0.15 \pm 0.02$	$0.96 \pm 0.05$
12	<i>P. adscendens</i>	$9.9 \pm 0.2$	$9.8 \pm 0.2$	$0.47 \pm 0.04$	$0.99 \pm 0.03$
14	<i>Squamarina</i> cfr <i>. stella-petraea</i> Poelt	$15.9 \pm 0.6$	$15.3 \pm 0.6$	$0.79 \pm 0.09$	$0.96 \pm 0.05$
15	<i>Ramalina fraxinea</i> (L.) Ach.	$1.11 \pm 0.05$	$1.11 \pm 0.05$	$0.05 \pm 0.01$	$1.00 \pm 0.06$
	<i>P. sulcata</i>	$3.4 \pm 0.1$	$3.4 \pm 0.1$	$0.19 \pm 0.02$	$1.01 \pm 0.04$
	<i>P. saxatilis</i>	$4.8 \pm 0.2$	$4.6 \pm 0.2$	$0.24 \pm 0.03$	$0.96 \pm 0.05$

(1) In these samples no separation between bark and lichen has been made.

The uncertainties reported in Table I include a) the uncertainty associated with weighing of the sample; b) the uncertainty associated with the activity of the tracer ( $^{232}\text{U}$ ), as well as the uncertainty associated with the addition of the tracer to the sample; and c) the uncertainty associated with the counting statistics of the sample and the blank.

The results indicate that  $^{235}\text{U}$  data have very high uncertainties. This is attributable to the low  $^{235}\text{U}$  activity concentrations measured in the biological samples. This isotope is more accurately estimated using mass-spectrometric techniques (Bou-Rabee, 1995) rather than by alpha pulse-height analysis. In this regard, the evaluation of the results has been made using only the  $^{238}\text{U}$  and  $^{234}\text{U}$  activity concentration data obtained by alpha-spectrometry because of the sensitivity of this method is suitable for such measurements (Sansone *et al.*, 2001a).

The inter-laboratory exercise carried out between APAT and Spiez laboratories using the Reference Material IAEA 336 Lichen (Freitas *et al.*, 1993), indicated a  $^{234}\text{U}/^{238}\text{U}$  mean activity ratio of  $0.95 \pm 0.08$  characteristic of natural uranium. On this basis, taking into account the lowest value of the intercomparison activity ratios ( $^{234}\text{U}/^{238}\text{U}$   $0.92 \pm 0.08$ ), it is possible to argue that  $^{234}\text{U}/^{238}\text{U}$  activity concentration ratio values (measured in botanical samples collected in Bosnia–Herzegovina) that are below 0.84, including the associated uncertainty, could be indicative of anthropogenic contributions of depleted uranium (Figure 2).

The high variability of  $^{238}\text{U}$  data reported in Table I could be attributed to various factors:

- the different geological background of the sampling sites; namely, U is usually present in variable amounts in rock and after resuspension of such particles, may be captured by lichens (Loppi *et al.*, 2003);
- the different levels of DU contamination at each sampling site;

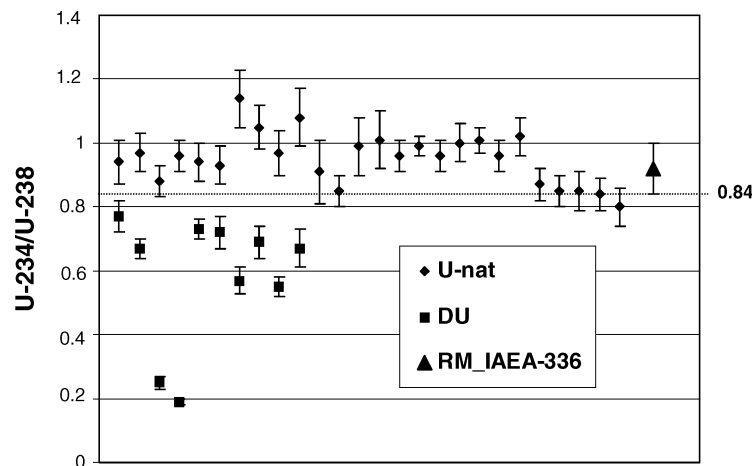


Figure 2. The  $^{234}\text{U}/^{238}\text{U}$  activity concentration ratios in botanical samples collected in Bosnia and Herzegovina. Ratios below 0.84 could be indicative for depleted uranium.

- the different exposure of lichen arising from to the position of the trees with respect to soil particles dispersed in the air, in relation to prevailing wind directions (Loppi *et al.*, 1999), and
- the different mechanisms of uranium bio-accumulation in different lichen species (McLean *et al.*, 1998; Di Lella *et al.*, 2003).

In the present samples collected in Bosnia–Herzegovina, the  $^{234}\text{U}/^{238}\text{U}$  activity ratios (Table I) range from  $0.19 \pm 0.01$  (site 4) to  $1.14 \pm 0.09$  (site 3). On the basis of the results of the inter-laboratory exercise, it is possible to discriminate 10 samples in three locations where DU is detectable (Figure 2): in two samples collected at the former *Hadzici Tank Repair Facility* (site 1), all samples collected at the *Hadzici Ammunition Storage Depot* (site 4) and in some samples from the *Han Pijesak Barracks* (site 7). During the war, these three sites were the main target for A-10 attack with DU ammunition and some places were totally destroyed (UNEP, 2003; Rosamilia *et al.*, 2003). In any case, most  $^{234}\text{U}/^{238}\text{U}$  activity ratios related to sites 1, 4 and 7 are close to values indicating the presence of depleted uranium, as reported in Figure 2. The remaining samples show  $^{234}\text{U}/^{238}\text{U}$  activity concentration ratios are indicative of natural uranium contributions.

The accumulation rate of uranium in lichens mainly depends on the exposed surface area of the lichen, which can be related to the type of reproduction and growth form (Di Lella *et al.*, 2003). Although lichens show different uptake abilities, several epiphytic species can be used as biomonitors, and *H. physodes*, *E. prunastri*, and different *Parmelia* species, which have been used during the present survey, are among the most frequently applied (Bargagli and Mikhailova, 2002; Jeran *et al.*, 1995; Kirchner and Daillant, 2002). The lichen species collected in sites

where the lowest  $^{234}\text{U}/^{238}\text{U}$  values were recorded are *P. sulcata*, *P. saxatilis*, and *H. physodes*.

### 3.2. METALS

Using  $k_0$ -INAA, the elements As, Ba, Ca, Co, Cr, Fe, Sb, Sc, Th, U, and Zn were quantitatively determined in lichen and bark samples. The results are given in Table II. Also in Table II the  $^{238}\text{U}$  levels in mg/kg obtained by recalculation of activity concentration of  $^{238}\text{U}$  (Bq/kg) found by alpha-spectrometry in the APAT laboratories are added. Although two different methods were used, namely  $k_0$ -INAA at IJS and alpha-spectrometry in APAT, a good agreement was found.

The results did not show any correlation between DU and metals. However, a high correlation between total uranium (or  $^{238}\text{U}$ ) was observed for As, Ba, Ca, Cr, Fe, Sb, Sc, and Th (Table III) if all lichen species from all locations investigated were taken into account.

Elevated amounts of metals in lichens are mostly ascribable to particles trapped within the thallus. The amount of metal accumulated by a lichen is species-dependent and refers decisively to its morphological structure (Chiarenzelli *et al.*, 2001; Di Lella *et al.*, 2003). For example, foliose lichens appear to have a higher Fe content than fruticose lichens. The difference between these growth forms was explained by the easy accessibility to soil and ultra fine rock debris of rock-inhabiting foliose species (Garty, 2001). In the case of *Squamarina* the high values could be attributed to the fact that this saxicolous specie could be affected by fine rock debris. Numerous studies have been performed on environmental pollution in the surroundings of contaminated sites which showed a decrease of metal content in in situ lichens sampled at increasing distance along transects from metal smelters, steelworks, power stations, mining sites, or mixed industrial centres from the source of emission (Jeran *et al.*, 1995; Nieboer *et al.*, 1982).

An explanation for the rather high variability of trace element contents in the lichens and tree barks collected might be that only part of the total content could be attributed to pollution, the other part representing the natural origin of the elements, where the particular geology of the territory should not be neglected (Jeran *et al.*, 2001).

For most elements the highest values were found in the samples collected at the Kalinovik Ammunition Destruction Site (site 14) and Bjelasnica Plateau site (site 15). These two sites have been used for many years to destroy ammunition/UXO. It is well known that elements such As, Co, Cr, Fe, Sb, and Zn are used in steel production so it is reasonable to find high values at these sites. Very high levels of Sc, Th, and U also indicate a soil contribution from explosion of the ammunition.

In the other sites, the element concentrations are in good agreement with other studies carried out in Slovenia (Jeran *et al.*, 2002) and in Montenegro (Jovanović *et al.*, 1995) in contaminated areas. In fact, all the sites where the highest metal

Table II. Concentrations of metal and other elements in lichen and tree Bark samples collected in Bosnia-Herzegovina

Sample type	Site	As (mg/kg)	Ba (mg/kg)	Ca (mg/kg)	Co (mg/kg)	Cr (mg/kg)	Fe (mg/kg)	Sb (mg/kg)	Sr (mg/kg)	Th (mg/kg)	U (mg/kg)	$^{238}\text{U}^*$ (mg/kg)	Zn (mg/kg)
<i>Hypogymnia physodes</i>	1	1.15 ± 0.04	39.8 ± 1.5	44060 ± 1543	0.59 ± 0.02	12.6 ± 0.4	1556 ± 55	0.48 ± 0.019	0.369 ± 0.013	0.26 ± 0.01	0.12 ± 0.005	0.15 ± 0.008	55 ± 1.9
(L.) Nyl.													
<i>Parmelia sulcata</i>	1	1.60 ± 0.06	41.5 ± 1.7	12540 ± 444	0.72 ± 0.03	15.7 ± 0.6	1703 ± 62	0.56 ± 0.023	0.399 ± 0.015	0.27 ± 0.01	0.16 ± 0.007	0.18 ± 0.007	66 ± 2.6
Taylor													
<i>P. sulcata</i>	1	1.16 ± 0.04	39.1 ± 1.4	17780 ± 623	0.57 ± 0.02	11.1 ± 0.4	1535 ± 54	0.36 ± 0.014	0.380 ± 0.014	0.28 ± 0.01	0.16 ± 0.007	0.18 ± 0.007	75 ± 2.6
<i>P. sulcata</i>	1	1.46 ± 0.05	47.4 ± 1.7	8322 ± 293	0.83 ± 0.03	14.7 ± 0.5	2002 ± 70	0.52 ± 0.020	0.383 ± 0.014	0.26 ± 0.01	0.30 ± 0.012	0.29 ± 0.010	69 ± 2.4
Bark	1	0.55 ± 0.02	110 ± 4	35820 ± 1255	0.50 ± 0.02	4.93 ± 0.17	387 ± 14	0.49 ± 0.018	0.103 ± 0.004	0.10 ± 0.004	0.06 ± 0.006	0.08 ± 0.003	27 ± 0.9
Bark and	1	0.40 ± 0.02	78.2 ± 2.8	30500 ± 1069	0.46 ± 0.02	3.63 ± 0.13	633 ± 22	0.21 ± 0.008	0.182 ± 0.006	0.17 ± 0.01	0.08 ± 0.009	0.09 ± 0.004	19 ± 0.7
<i>Arthopyrenia</i>													
sp.													
Bark and	1	1.04 ± 0.04	230 ± 8	39730 ± 1400	0.72 ± 0.03	6.24 ± 0.22	821 ± 29	0.38 ± 0.014	0.267 ± 0.009	0.29 ± 0.02	0.13 ± 0.006	0.14 ± 0.006	153 ± 5
<i>Arthopyrenia</i>													
sp.													
Bark	2	0.33 ± 0.02	19.7 ± 0.8	30880 ± 1089	0.56 ± 0.02	49.8 ± 1.7	724 ± 25	0.24 ± 0.009	0.161 ± 0.006	0.17 ± 0.01	0.07 ± 0.003	0.07 ± 0.003	19 ± 0.7
<i>P. sulcata</i>	2	1.58 ± 0.06	39.3 ± 1.7	19340 ± 686	0.90 ± 0.03	10.3 ± 0.4	2123 ± 74	0.56 ± 0.020	0.637 ± 0.026	0.57 ± 0.02	0.19 ± 0.009	0.19 ± 0.007	130 ± 5
<i>Physcia</i>	2	2.22 ± 0.08	39.9 ± 1.8	19690 ± 714	1.05 ± 0.04	15.5 ± 0.5	2971 ± 104	0.61 ± 0.022	0.812 ± 0.029	0.75 ± 0.03	0.22 ± 0.009	0.24 ± 0.011	117 ± 4
<i>adscendens</i>													
Fr. H. Oliver													
<i>P. sulcata</i>	3	1.07 ± 0.04	21.6 ± 0.9	7645 ± 278	0.68 ± 0.03	5.64 ± 0.20	1608 ± 56	0.40 ± 0.015	0.491 ± 0.017	0.38 ± 0.01	0.13 ± 0.008	0.15 ± 0.007	52 ± 1.8
Bark	4	nd	14.3 ± 0.6	31170 ± 1109	0.07 ± 0.00	3.66 ± 0.13	96.6 ± 3.5	0.01 ± 0.001	0.022 ± 0.001	0.02 ± 0.001	nd	0.03 ± 0.002	9 ± 0.36
<i>H. physodes</i>	6	0.69 ± 0.03	35.4 ± 1.4	29160 ± 1029	0.58 ± 0.02	8.08 ± 0.28	1174 ± 41	0.21 ± 0.008	0.384 ± 0.014	0.28 ± 0.01	0.09 ± 0.015	0.10 ± 0.005	57 ± 2.0

<i>P. sulcata</i>	6	1.35 ± 0.05	60.9 ± 2.3	7413 ± 274	0.67 ± 0.03	10.5 ± 0.4	1758 ± 62	0.39 ± 0.017	0.542 ± 0.019	0.39 ± 0.01	0.17 ± 0.008	0.16 ± 0.008	60 ± 2.1
<i>H. physodes</i>	6	0.93 ± 0.04	62.8 ± 2.3	25070 ± 879	0.45 ± 0.02	5.60 ± 0.20	1121 ± 39	0.38 ± 0.014	0.319 ± 0.011	0.22 ± 0.01	0.09 ± 0.006	0.10 ± 0.006	51 ± 1.8
<i>P. sulcata</i>	7	1.58 ± 0.06	25.3 ± 1.2	11070 ± 392	0.98 ± 0.04	6.41 ± 0.23	1471 ± 52	0.25 ± 0.009	0.426 ± 0.015	0.37 ± 0.01	0.19 ± 0.008	0.22 ± 0.007	42 ± 1.5
<i>Platismatia glauca</i>	7	1.43 ± 0.05	19.0 ± 0.8	3036 ± 109	0.35 ± 0.01	4.38 ± 0.16	727 ± 26	0.37 ± 0.013	0.249 ± 0.009	0.21 ± 0.01	0.09 ± 0.004	0.10 ± 0.004	39 ± 1.4
(L.) W.L. Culb. and C.F. Culb.													
<i>H. physodes</i>	7	0.52 ± 0.03	14.4 ± 0.6	5442 ± 192	0.28 ± 0.01	3.16 ± 0.11	626 ± 22	0.18 ± 0.007	0.184 ± 0.006	0.16 ± 0.01	0.12 ± 0.006	0.11 ± 0.005	51 ± 1.8
<i>H. physodes</i>	7	0.76 ± 0.03	16.3 ± 0.9	10520 ± 372	0.29 ± 0.01	2.27 ± 0.08	769 ± 27	0.21 ± 0.008	0.216 ± 0.008	0.20 ± 0.01	0.11 ± 0.017	0.11 ± 0.005	52 ± 1.8
Bark	10	0.97 ± 0.04	43.7 ± 1.6	25230 ± 888	0.56 ± 0.02	9.66 ± 0.34	1308 ± 46	1.00 ± 0.04	0.236 ± 0.008	0.15 ± 0.01	0.10 ± 0.013	0.10 ± 0.004	50 ± 1.8
<i>H. physodes</i>	10	0.55 ± 0.03	19.1 ± 0.9	24350 ± 853	0.46 ± 0.02	3.98 ± 0.14	1160 ± 41	0.33 ± 0.012	0.343 ± 0.012	0.27 ± 0.01	0.07 ± 0.003	0.07 ± 0.005	47 ± 1.7
<i>Xanthoria parietina</i>	10	1.89 ± 0.07	50.9 ± 1.9	7117 ± 283	1.23 ± 0.04	10.3 ± 0.4	3314 ± 116	0.60 ± 0.03	1.08 ± 0.04	0.82 ± 0.03	0.21 ± 0.010	0.21 ± 0.007	39 ± 1.4
(L.) Th.Fr.													
<i>P. adscendens</i>	12	0.38 ± 0.02	26.7 ± 1.1	21870 ± 768	0.66 ± 0.03	6.27 ± 0.22	923 ± 33	0.13 ± 0.007	0.266 ± 0.009	0.22 ± 0.01	0.12 ± 0.005	0.08 ± 0.002	378 ± 13

\*Determined by alpha-spectrometry.

nd: not detectable.

Table III. Correlations between metals and  $^{238}\text{U}$  in lichen samples (N = 18)

	As*	Ba*	Ca*	Cr*	Fe*	Sb*	Sc*	Th*	U*	Zn
U-238	0.93	0.66	0.82	0.98	0.98	0.97	0.97	0.98	1.00	0.28

Correlations marked with an asterisk are significant at  $p < 0.05$ .

values were found are areas where anthropogenic activity, sometimes associated with urban traffic, is very high.

#### 4. Conclusions

In conclusion, the main outcome of this investigation is that lichens and tree barks are sensitive bio-accumulators of past airborne contamination by depleted uranium dusts or aerosol particles generated at the time of the attack. At three heavily bombed sites, the presence of DU in lichen and tree bark samples indicates the earlier presence of DU in the air, which implies that, more than 9 years ago, at least some of the penetrators hit hard targets and surfaces and shattered into dust and dispersed into the air.

- The presence of DU in lichen and tree/bush bark is mainly attributable to:
- direct deposition of DU dust particles during the attack;
- deposition of suspended materials in air (originating from resuspension of soil and deposited DU dust particles).

The trace element analysis provided information about possible high concentrations in the investigated sites determined by anthropogenic activities. In particular, the results showed significantly higher concentrations of most metals at sites used for ammunition destruction. Further studies at these sites are necessary to evaluate the real contribution of civil activity to heavy metal pollution.

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