RADIOCHEMICAL AND CHEMICAL CONTAMINATION OF UNIMPROVED ECOSYSTEMS IN YUGOSLAVIA

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ABSTRACT

In this work we investigated the contamination of the following mountain ecosystems: Sinjajevina, Durmitor, Bjelasica, Seličevica, Tara and Kopaonik, by analyzing the activity levels of ^{137}Cs and content of heavy metals (Pb, Cu, Zn, Ni, Cr, Cd) in bioindicator organisms (lichens, mosses and mushrooms). High activity levels of ^{137}Cs remained in all bioindicator species, although the accident at the nuclear power plant in Chernobyl happened 15 years ago in 1986. Heavy metals contents were low in nearly all samples of lichens, mosses and mushrooms. Only one high result was found for the content of lead in moss originating from Kopaonik mountain.

Key words: unimproved ecosystems, radiochemical contamination, chemical contamination, bioindicators

INTRODUCTION

Highland ecosystems occupys very little space in Serbia, even though that Yugoslavia contains some of the highest land in Europe. However, although 57% of the territory in Montenegro is above 1000 m altitude, Serbia has 11% and only 0,5% of the territory is above 2000 m altitude in the whole country. These highest parts are distributed irregularly and they are located around mountain peaks (l).

The mountain ecosystems in Yugoslavia can be divided into two groups from the aspect of the natural conditions of the habitats and their organisms, i e: European type (Zlatar, Zlatibor, Golija, Kopaonik, Stara planina, Suva planina) and South Balkan type (Šar planina and Prokletije).

These highland ecosystems are considered to be very sensitive, because of their slow autoreparation. The relationships in the ecosystems reestablish by long term processes of interaction existence with short periods of specific climatic conditions and short periods of vegetation and production.

The permanent value of the mountain ecosystems is their biological energetic potential (food and animal feed), which is the most important for the local inhabitants.
Environmental contamination of these regions, especially of ruminants is a more serious problem than of the lowland agricultural lands.
Unimproved ecosystems provide relatively small amounts of food, but the total radiation dose for the inhabitants can be very high, as was the case after the accident at the nuclear power plant in Chernobyl on April 26, 1986. Wooded complexes at higher altitudes are predisposed to receive more fallout and then more radionuclides. For example, Norwegian authors found 90000 $Bqkg^{-1}$ of ^{137}Cs in deer meat in 1988 (2).
Meat and milk products from ruminants grazing unimproved ecosystems are the most common direct route by which radioactive contamination in these areas reaches the human food chain, although fish, wildfowl, fungi, and berries can contribute significantly in certain regions.
Countermeasures have been developed and used succesfully to reduce radiocesium levels in ruminants grazing in unimproved ecosystems. Apart from decontamination by altering farming practices and providing uncontaminated feeds, sustained reductions of 50% to 80% in the radiocesium concentrations of both milk and meat have been achieved in many ruminant species when AFCF (ammonium iron hexacyanoferrate) is given via a sodium chloride lick or as a sustained-release bolus. Food production in unimproved ecosystems must be evaluated separately from that of ordinary agricultural systems.
Contamination of highland ecosystems was described by A. Stanković et al., including results for ^{137}Cs activity levels in food and animal feed, as well as lichens and mosses as bioindicator species, which were collected on Durmitor mountain in 1993 (3).
Activity levels of ^{137}Cs and concentrations of heavy metals (Pb, Cu, Zn, Ni, Cr, Cd) were analysed in some bioindicator species collected on two Serbian mountains (Tara and Kopaonik), which are Yugoslav National Parks (4).
All these bioindicator species are biomonitors for the environmental pollution. Biomonitoring, in general sense, is the use of properties of an organism or a part of it to obtain information on a certain part of the biosphere.
The relevant information in biomonitoring programmes using plants or animals is commonly deduced from either changes in the behaviour of the monitor organism (species composition, -richness, ecological performance, morphology) or from the concentrations of specific substances in the monitor tissues (5).
To be suitable for application as monitor for air particulate matter, specific requirements have to be met by a biological tissue or monitor. General criteria of primary importance are:

a) the response of the organism to the quantity of the elements to be monitored should be known
b) the organisms should be common in the area of interest
c) availability is required at any time or season
d) the monitor should be tolerant to pollutants at relevant levels
e) the element uptake should not be influenced by regulating biological mechanisms or antagonistic or synergistic effects
f) the biomonitor should average the element concentrations over a suitable time period as a result of integrated exposure over a time period
g) the accumulation should reach to concentration levels which are accessible by routine analytical techniques.

The morphology of lichens and mosses does not vary with seasons, thus accumulation can occur throughout the year. Lichens and mosses usually have considerable longevity, which led to their use as long-term integrators of atmospheric deposition (6). Thus, tolerant lichen species and mosses are likely to be the best suitable organisms.

In this article we investigated the contamination of some mountain ecosystems by analyzing the activity levels of ^{137}Cs and contents of heavy metals (Pb, Cu, Zn, Ni, Cr, Cd) only in the bioindicator organisms (lichens, mosses and mushrooms) which have the property to represent average concentrations of radionuclides and heavy metals as the result of integral exposure in the examined period of time (7).

In many cases both pre-Chernobyl and Chernobyl radiocesium levels have now equilibrated with stable cesium in these ecosystems. It is therefore likely that future changes in radiocesium activities in vegetation will be determined by a combination of the physical and ecological half-life of the radiocesium isotopes and not by the initial physicochemical properties of the deposition. It seems appropriate to review advances in our understanding of the long-term behavior of radiocesium in unimproved ecosystems (8).

EXPERIMENTS AND RESULTS

All samples of bioindicators (lichens, mosses and mushrooms) were collected in Serbia and Montenegro, immediately after the accident in Chernobyl and then continualy till now. After homogenization they were measured in Marinelli beakers by the gammaspectrometry method using an ORTEC-CANBERRA gammaspectrometer with 8192 channels (resolution 6,8% for ^{137}Cs) and at a counting efficiency of 8,7%. In order to determine concentrations of heavy metals, samples (0.5g) were destructed with nitric acid (concentrated) and hydrogen-peroxyde. Solutions were filtered and then analysed for Pb, Cu, Zn, Ni, Cr and Cd by atomic absorption spectrophotometry (spectrophotometer PYE UNICAM SP-192).

The activity levels of ^{137}Cs in lichens, mosses and mushrooms collected in Montenegro after the accident in Chernobyl are presented in Table I.

It is obvious that ^{137}Cs activity levels were the highest just after the accident in Chernobyl, then the radioactivity levels decreased due to physical decay and biological loss. A. Stanković was determined the biological and effective half life for the lichen species *Evernia prunastri* collected on Durmitor mountain 1991.(9), by applying the next three equations (10).

$$A_t = A_0\, e^{-(\lambda+\lambda b)t} \qquad /1/$$

where A_0 is the radioactivity of the sample at t=0,

A_t is the the radioactivity of the sample at $t \neq 0$.

λ is the radioactivity decay constant = $\ln2/T_f = 0{,}023$ year $^{-1}$

T_f is the physical half life,

T_b is the biological half life

$$\Lambda_b = \ln2/T_b \qquad /2/$$

Using the equation /2/ it was possible to calculate the biological half life T_b, and finally, the effective half life T_{eff} was calculated by applying the next equation:

$$T_{eff} = T_f\, T_b/ (T_f + T_b) \qquad /3/$$

Lichen sample was measuring successively three years during the anabiosis state of plant.

The values for the T_{eff} were between 6,7 and 7,5 years, but for the T_b results were in the interval from 8,6 to 10,1 years for the lichen species *Evernia prunastri* .

These values are dependent on the chemical type of radionuclide, pH of the medium, its temperature, and the type of the lichen species.

Table I The activity levels of ^{137}Cs in lichens, mosses and mushrooms collected in Montenegro

Sample	Location	Year	^{137}Cs (Bqkg^{-1})
Lichens			
Evernia prunastri	Sinjajevina	1986	7100 ± 700
Evernia prunastri		1987	12000 ± 1000
Evernia prunastri		1988	9500 ± 900
Evernia prunastri		1989	6500 ± 700
Evernia prunastri		1990	3600 ± 400
Evernia prunastri		1991	4000 ± 350
Evernia prunastri		1992	5500 ± 600
Usnea barbata	Durmitor	1990	2700 ± 300
Usnea barbata		1991	2400 ± 250
Usnea barbata		1992	2250 ± 200
Usnea barbata		1993	1000 ± 100
Usnea barbata		1994	500 ± 50
Usnea barbata		1995	250 ± 30
Pseudoevernia furfuracea		1993	1600 ± 200
Peltigera canina		1993	2300 ± 250
Certraria islandica		1993	1100 ± 150
Cladonia fimbriata		1993	3250 ± 350
Mosses			
Plagiothecium sp.	Durmitor	1993	3500 ± 400
Plagiothecium sp.		1993	5181 ± 600
Homalothecium sericeum	Bjelasica	2000	1005 ± 100
Mushrooms			
Cantharellus cibarius	Durmitor	1986	100 ± 10
Morchella conica		1986	1200 ± 120
Boletus edulis		1986	50 ± 5
Morchella conica		1987	580 ± 60
Morchella conica		1988	820 ± 80
Morchella conica		1990	400 ± 40

Table II shows the activity levels of ^{137}Cs in lichens, mosses and mushrooms collected in Serbia during 1996 and 2000.

There was lower activity of ^{137}Cs in these samples compared with the activity levels in those from Montenegro (Table I), especially in lichen species collected on Seličevica mountain in 1986. The highest value for ^{137}Cs activity was measured on Tara mountain in 2000. in lichen type *Peltigera polydactila* (5801±600) Bq/kg, which is the result of the high altitude of the place where lichen was growing. Comparing all data about ^{137}Cs activity levels it can be concluded that the content species was mostly depending on the type of bioindicator and the altitude of the mountain system.

However, it is evident that ^{137}Cs is still present in all types of bioindicators analyzed in this work, although the accident at the nuclear power plant in Chernobyl happened fifteen years ago.

Table II The activity levels of ^{137}Cs in lichens, mosses and mushrooms collected in Serbia

Sample	Location	Year	^{137}Cs (Bqkg^{-1})
Lichens			
Cladonia foliacea (Hudson)	Seličevica	1986	345 ± 40
Cladonia rangiformis Hoffm		1986	110 ± 10
Cladonia fimbriata (L.) Fr		1986	2336 ± 50
Peltigera polydactila (Necker)	Tara	2000	5801 ± 600
Cladonia furcata (Hudson)		2000	576 ± 60
Pseudoevernia furfuracea (L.)		2000	688 ± 70
Pseudoevernia furfuracea (L.)	Kopaonik	2000	776 ± 80
Certraria islandica (L.) Ach.		2000	512 ± 50
Usnea florida (L.) Web. In		2000	274 ± 30
Bryoria fuscescens (Gyeln.)		2000	383 ± 40
Mosses			
Homalothecium sericeum	Tara	2000	896 ± 90
Polytrichum juniperinum Willd		2000	808 ± 80
Hypnum cupressiforme Hedw.		2000	522 ± 50
Tortula subulata Hedw.	Kopaonik	2000	3646 ± 400
Hypnum cupressiforme Hedw.		2000	2354 ± 250
Polytrichum juniperinum Hedw		2000	1776 ± 180
Pseudoscleropodium purum		2000	1405 ± 140
Sphagnum fuscum (Schimp.)		2000	1011 ± 10
Mushrooms			
Boletus edulis (dry)	Zapadno Pomoravlje	1999	77 ± 21
Cantharellus cibarius (dry)		1999	100 ± 38
H. repandum (fresh)		1999	205 ± 85
Morchella conica (dry)		1999	134 ± 20
Boletus edulis fresh		2000	190 ± 35
Cantharellus cibarius		2000	141 ± 35
C. cornicopioides		2000	111 ± 20

The results presented in Table III show the lower values for the contents of heavy metals nearly in all samples examined compared with the action levels. Only one sample shows higher value for the content of lead in moss (Kopaonik), probably affected by emission of Trepča complex.

CONCLUSION

Our results show high activity levels of ^{137}Cs remaining in all bioindicator species examined in this work. Radioactivity monitoring for these samples is very important because the possible contamination of the food chain, and finally to decrease the radiation body burden of the inhabitants.

Table III Concetrations of some heavy metals in lichens, mosses and mushrooms collected in Serbia

Sample	Location	Concentration of metals (ppm)					
		Pb	Cu	Zn	Ni	Cr	Cd
Lichens							
Peltigera polydactila (Necker)	Tara	38,1	5,9	37,1	0,1	0,7	0,1
Cladonia furcata (Hudson)		26,2	8,0	15,0	0,1	1,0	0,1
Pseudoevernia furfuracea (L.)		8,9	5,3	6,3	0,3	0,7	0,1
Pseudoevernia furfuracea (L.)	Kopaonik	11,4	17,1	5,8	0,2	2,0	0,1
Certraria islandica (L.) Ach.		11,2	20,0	1,1	0,4	2,0	0,1
Usnea florida (L.) Web. In		33,1	43,1	58,3	3,6	0,5	0,2
Bryoria fuscescens (Gyeln.)		2,0	39,8	6,0	0,2	1,2	0,1
Mosses							
Homalothecium sericeum	Tara	6,3	6,1	3,8	0,1	1,8	0,1
Tortula subulata Hedw.	Kopaonik	96,2	38,7	58,1	0,2	1,1	0,3
Hypnum cupressiforme Hedw.		133,	38,3	54,3	5,4	2,0	0,6
Polytrichum juniperinum Hedw		19,8	48,1	40,0	4,7	2,2	0,1
Pseudoscleropodium purum		50,6	44,1	45,0	5,8	3,9	0,2
Sphagnum fuscum (Schimp.)		3,1	20,2	9,1	0,1	3,1	0,1
Mushrooms							
B. edulis	Zapadno Pomoravlje	46	23	-	-	2,1	0,1
C. cibarius		69	57	-	-	1,9	0,2
C. cornicopioides		58	65	-	-	1,4	0,1

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