Measurements on Chernobyl Fallout in Forest Vegetation*

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Forests play a special role in the general problem of the ionizing radiation effects on the biosphere. Interest in the biogeophysical consequences of forest irradiation depends on their extreme susceptibility to radioactive contamination, owing to the high radiosensitivity of woody plants, and to the pronounced interceptive capacity of their canopies with respect to radioactive fallout.

Three months after the Chernobyl accident the direct contamination of components of forest cenoses. located in a typical mountain area of northern Italy (Auronzo, Dolomites: 850-1200 m above sea level) was investigated. The following radionuclides were identified: 134 Cs, 137 Cs, 103 Ru, 106 Rh $^{-106}$ Ru, 141 Ce, 144 Ce, 140 Ba $^{-140}$ La, 95 Zr $^{-95}$ Nb, 125 Sb, 110m Ag.

The aim of this study was to measure the rare-earth radionuclides ¹⁴¹Ce, ¹⁴⁴Ce and ¹⁴⁰Ba-¹⁴⁰La related to ¹³⁷Cs, which, using the words in much the same sense as the International Commission on Radiological Protection [1], can be appropriately described as 'critical nuclide', being a main source of radiation to all members of the community.

Materials and Methods

On July 10th. 1986 different above-ground organs of the following forest species were sampled: spruce (Picea abies L.) and scotch pine (Pinus silvestris L.), both evergreen; larch (Larix decidua, Mill.). Lichens (Usmea florida L.) were collected from spruce branches. All these organic materials were oven-dried at 105 °C to constant weight, pulverized, homogenized and analyzed by gamma-spectrometry, using a 140 cm³ PGT coaxial intrinsic germanium detector: efficiency relative to 1.33 MeV ⁶⁰Co gamma-ray efficiency obtained with a $3'' \times 3''$ Nal(Tl) detector at a distance of 25 cm: 30.7%; energy resolution (full width at half maximum): 2.02 keV at 1.33 MeV; shielding: 10 cm lead, 0.1 cm lead, 0.1 cm cadmium, 0.1 cm copper, 0.8 cm methacrylic resin. The detector was coupled with a 8K Cicero-Silena analyzer; the peak analysis was performed using a Silenaquantitative isotopic analysis option, mod. 8500-Quan. Counts were made on 250 cm³ Marinelli beakers for 500–1200 min.

The counting efficiency in the sample geometry was determined for different photon energies from measurements on samples containing known amounts of gamma-ray reference solution QCy 44, Amersham. This solution was mixed with 'before Chernobyl' forage samples to simulate the density and the atomic number of plant material.

Data (expressed in Bq/kg dry weight) are affected by statistical counting errors ranging from $\pm 6\%$ to 25% at a 95% confidence level. The uncertainty due to the sampling effects and to biological variations was not determined, but, according to the sampling criteria, it was estimated as reasonably low.

Experimental data are listed in Table I.

Results and Conclusions

The rare-earth radionuclides ¹⁴¹Ce, ¹⁴⁴Ce, ¹⁴⁰Ba-¹⁴⁰La as components of radioactive fallout are normally considered devoid of importance from the point of the radiological health, because they are supposed to enter food chains to only a very small extent. But, on account of a relatively long half-life and ability to be deposited in the skeleton bones, if absorbed in the body, their presence in the environment cannot be ignored.

The samples examined are organs of conifers which, according to Sparrow [2], are among the most 'critical members' of woodland ecosystems because they are extremely sensitive to radiocontamination.

Radionuclides primarily intercepted on aboveground organs of the plants can afterwards migrate to the internal tissues or are removed from plants by rain, wind, etc. The mechanism of foliar absorption has been regarded as largely outside the aim of this investigation.

In Table II the activity ratios, decay corrected to April 29, 1986 and normalized to ¹³⁷Cs, are listed for the tree organs reported in Table I. The nuclide inventory of the Chernobyl-4 reactor, the percentage released after the accident along with the ratios in the core, normalized to ¹³⁷Cs, are given approximately in Table III [3, 4]. In our experiments only the ratio

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^{*}Paper presented at the Second International Conference on the Basic and Applied Chemistry of f-Transition (Lanthanide and Actinide) and Related Elements (2nd ICLA), Lisbon, Portugal, April 6-10, 1987.

Ecosystems	Samples	Radionuclides			
		137Cs	¹⁴¹ Ce	¹⁴⁴ Ce	¹⁴⁰ Ba- ¹⁴⁰ La
Spruce ^d stand	(old ^f	1870	71	230	<lda<sup>c</lda<sup>
	voung	340	<lda< td=""><td><26</td><td><lda< td=""></lda<></td></lda<>	<26	<lda< td=""></lda<>
	twigs $(1-3 \text{ years})$	3750	144	420	243
	cones (old)	610	17	49	<lda< td=""></lda<>
	lichens	12100	354	1040	400
Scotch pine ^d stand	., (old ^f	2150	97	262	39
	needles young	370	<7	<21	<lda< td=""></lda<>
	twigs (1-3 years)	2520	123	325	80
Larch ^e stand	twigs (1-3 years)	1600	26	76	<lda< td=""></lda<>
	needles (young)	1360	33	104	<29
	cones (young)	428	<25	<34	<lda< td=""></lda<>

TABLE I. Radioactivity (Bq/kg d.w.^a) Detected on July 10, 1986 in Different Ecosystems of Auronzo Forest Area (Dolomites, Italy)^b

^aDry weight. ^bData performed with uncertainty of ±6% to 25%; the symbol < means an uncertainty exceeding 25%. ^cLDA (lowest detectable activity) 0.9 Bq/kg d.w. at 95% confidence level for a counting time of 1200 min. ^dEvergreen. ^eDeciduous. ^f2-3 Years.

TABLE II. Activity Ratios (Bq/kg d.w.) Decay Corrected to April 29, 1986, and Normalized to ¹³⁷Cs for some Organs Listed in Table I

Species	Organs	Radionuclides			
		¹³⁷ Cs	¹⁴¹ Ce	¹⁴⁴ Ce	¹⁴⁰ Ba- ¹⁴⁰ La
Spruce	needles (old)	1	0.18	0.14	a
	twigs (1-3 years)	1	0.18	0.13	3.4
	cones (old)	1	0.13	0.09	-
Scotch pine	needles (old)	1	0.21	0.14	0.9
	twigs (1-3 years)	1	0.23	0.15	-
Larch	twigs (1-3 years)	1	0.11	0.09	-
Lichens	thallus	1	0.13	0.10	1.7

^aSymbol – means that ratio cannot be calculated because nuclide activity is lower than LDA or affected by uncertainty >25%.

TABLE III. Nuclide Inventory, Percentage Released and Ratios in Core Normalized to $^{137}\mathrm{Cs}$ of the Chernobyl-4 Reactor

Nuclide	Inventory (Bq) $\times 10^{-18}$	Percentage released (%)	Ratios in core
¹³⁷ Cs	0.29	13	1
¹⁴¹ Ce	4.4	2.3	15
¹⁴⁴ Ce	3.2	2.8	11
¹⁴⁰ Ba- ¹⁴⁰ La	2.9	5.6	10

TABLE IV. Normal Melting and Boiling Points of Cesium, Lanthanide Elements and Oxides

Elements or oxide	Melting point (°C)	Boiling point (°C)
Cs	28.6	670
Ba	725	1640
Ce	799	3426
La	921	3457
Ce_2O_3	1692	
BaO	1918	2000

¹⁴¹Ce/¹⁴⁴Ce substantially agrees with the same ratio processed from Table III.

We believe that the trend in radiochemical composition of fallout can be explained on the basis of the normal melting and boiling points of cesium, lanthanide elements and oxides (Table IV) [3] and the temperature of the reactor core. If we assign 2000 $^{\circ}$ C to the core temperature, Cs and Ba are identified

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Fig. 1. ¹⁴⁴Ce activity of old organs of forest trees normalized to lichens assumed to be a 'biological indicator' of nuclear contamination.

as volatiles, Ce and La as refractories. If some oxidation of the nuclides is involved, Ba might behave as refractory.

Lichens are symbiotic organisms composed of fungi and algae. They grow very slowly and have a wide specific surface; they can effectively retain remarkable amounts of radionuclides. This could make it possible to use them as 'biological indicators' of the radioactive contamination of a region [5, 6] (Fig. 1).

References

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