Accumulation of cesium and strontium from contaminated soils by some "bioenergy" crop varieties

A.J. Grebenkov, S.F. Timofejev[1] and H. Vandenhove[2]

Institute of Power Engineering Problems, 99 Akademik Krasin Str., Sosny, 220109 Minsk, Belarus
[1] Research Institute of Radiology, 16 Feduninskogo Str., 246050 Gomel, Belarus
[2] Belgian Nuclear Research Centre, SCK-CEN, 200 Boeretang, 2400 Mol, Belgium

Abstract. One of the options of contaminated site remediation strategy is culturing the crop varieties, which have biomass yield high enough to be further used as a local fuel in contaminated regions. A number of field tests of two "bioenergy" crops, namely *Brassica juncea* and *Salix viminalis* was carried out. The study was focused on accumulation ability of the given varieties in extracting ^{90}Sr and ^{137}Cs from contaminated soil. Two types of soil, peaty and sandy, were examined. For *Brassica juncea*, the accumulation factor of ^{90}Sr was about two orders of magnitude higher than that of ^{137}Cs. For *Salix viminalis*, leaves had twofold higher capability to accumulate ^{90}Sr and more than threefold higher capability to accumulate ^{137}Cs compared to stems. In our example, the peaty soil contained higher content of exchangeable forms of Ca and K compared to the sandy soil. This was a reason that radionuclide accumulation factors were higher for willow grown on a sandy soil. It was also revealed that ratio between exchangeable and total forms of both radionuclides was relatively high in sandy soil, and this also caused the higher accumulation factor for willow grown on a sandy soil type compared to a peaty one.

1. INTRODUCTION

In Belarus, more than 1.8 million hectares of agricultural lands were exposed to radioactive fallout, and almost 20% of them have been excluded from economic turnover. Today, 15 years after Chernobyl Accident, economical and environmental remediation of contaminated farmlands become a key problem. Any countermeasure envisages either treatment of contaminated soil that mitigates negative effect of radionuclides on food chain, or creation of favorable conditions for safe reuse of contaminated lands with culturing the nonfood market oriented products. As to the later, the replacement of food crop species by some technical crops is exercised now. One of the options that could be successfully applied to agricultural practice is culturing the crop varieties, which have biomass yield high enough to be further used as a local fuel in contaminated regions. Despite the fact that such crops are not a part of human food chain, they nevertheless come within restriction that stipulates their use as a fuel.

In the present study, we carried out a number of field tests of two "bioenergy" crops, namely *Brassica juncea* and *Salix viminalis*. In addition to investigation of some parameters important for biomass-to-energy conversion, i.e. growing conditions and yield, the study was focused on accumulation ability of the given varieties in extracting ^{90}Sr and ^{137}Cs from contaminated soil. Along with our data already reported in [1, 2], the results of the given study can be used for assessment of radiation risk from application of contaminated biomass in conversion processes that will further allow evaluating feasibility of bioenergy production from the identified crop varieties grown in affected regions.

2. MATERIALS AND METHODS

2.1. Willow

In May 1997, four plantations of willow were established at Savichy Research Base and Masany Research Station in Gomel Province within 30-km zone of Chernobyl Power Plant. In each plantation, four willow clones were planted with fourfold replication. The clones are Rapp, Orm and Jorr (*Salix viminalis*), and Bjorn (*S. viminalis* x *S. schwerinii*). The general parameters of the plantations are presented in Table 1.

Table 1. Parameters of willow plantations

Location and field #	Landscape and soil type	Planting date	Area (ha)	Density (cuttings/ha)	Contamination (kBq/m^2)	
					^{137}Cs	^{90}Sr
Savichy, Field #1	Poorly drained dry meadow, Soddy-podzolic sandy on glacial sand superseded with loose sand	05/05/97	0.26	18000	1427±94	167±14
Savichy, Field #2	Lowland, Peaty-marsh on medium-thick well decomposed sedge-reed-woody peat	04/05/97	0.30	18000	12876±2275	1368±122
Masany, Field #3	Former arable land, Soddy-podzolic, sandy-loam based on alluvial gleic shallow-grainy sand	09/05/97	0.12	18000	-	-
Masany, Field #4	Wetland, Peat bog of lowland type	08/05/97	0.10	18000	2857±365	2659±1120

It has to be noted that growth of the willow plantations established on the both sandy fields was not successful. Willow in Masany sandy soil died almost all within the first growing season. In Savichy sandy soil, the plantation was a bit more successful and has survived with mortality rate of about 40%.

During the first cutting-back, willow biomass and soil matter was sampled to determine radionuclides content and principal nutrient concentration. Before measurements, eight soil samples from each test field were taken with 25-cm cores, homogenized, oven-dried and sieved. Stems and leaves of a willow plant (10 samples per each variety plot) were sampled separately, oven-dried, ground, homogenized, and two samples from the homogenized mixture were taken for further measurements.

2.2. Rape

In August 1997, the test field of *Brassica juncea* was established in exclusion zone nearby Savichy Research Base, Gomel Province. Soil type is a soddy-podzolic, loose loamy sandy soil on glacial associated sand superseded with loose sand. The test field was formed with plots to provide fivefold repetitive scheme. The size of each plot was 5.0m X 2.5m. Sowing was carried out manually with density of 7 kg/ha, the distance between rows being 30 cm. Tendance of the plantation was provided manually, including watering, weeding and clearing.

The main feature of soil was acidity coupled with low content of organic matter and mineral components (Table 2). The doses of lime fertilizer (dolomite powder) were estimated according to value of pH. One third of the dose was introduced while ploughing, and the balance was introduced before sowing. The optimal value of pH=6.0 was achieved by adding of $CaCO_3$. The doses of mineral fertilizers were evaluated according to norms taking into account the yield of mineral components with harvest assuming the later to be normally about 2000 kg of seeds per hectare.

The sampling of biomass and soil was performed at a rosette phase when the yield of rape from the test field achieved 0.68-0.81 kg/m^2 (green mass).

Table 2. Rape test field main characteristics

Test plot #	pH	Total C, %	Exchangeable forms, mg-eq/100g			Contamination (kBq/m^2)	
			K	Ca	Mg	^{137}Cs	^{90}Sr
1	4.34	0.91	0.13	1.79	0.25	426	45.5
3	4.10	0.85	0.17	1.59	0.30	531	41.8
5	4.32	0.93	0.27	1.71	0.21	430	42.9
7	4.54	0.87	0.22	1.80	0.39	332	45.5
9	4.41	0.90	0.20	1.74	0.41	397	54.4
Average	4.34	0.89	0.20	1.73	0.31	423±72	46.0±4.9

2.3. Analysis of Samples

The permanent and time dependent soil characteristics were defined with atomic-absorption analysis. To provide elemental analysis of the samples the ICP PLASMA spectroscope was used. Exchangeable forms of the measured nutrients and radionuclides were extracted with 1N acetous ammonia.

Content of ^{137}Cs in the samples was measured using a Ge(Li) detector system with a NaI(Tl) annulus. Two spectrometers were used, i.e. ADCAM-300 and NOKIA LP-4900.

Content of ^{90}Sr in the soil samples was determined by means of radiochemical method. The probe is formed from the samples ashed with HCl acid solution of 6M during 6 hours at 600°C followed by washing in ammonia solution at pH=8. Strontium is extracted in a form of carbonates. For plant matter, the probe is formed by ashing with concentrated HCl acid solution until forming oxalates during 7-20 hours at 500-600°C followed by transferring oxalates into their oxides at 700°C and cleaning against other radionuclides. The daughter isotope, ^{90}Y is extracted in a form of its oxalate and counted after 14 days. The chemical yield of ^{90}Sr is determined by an atomic adsorption spectrometer, while ^{90}Y is measured by weighing. Measurements of the targets were provided by Beta-spectrometer CANBERRA-2400.

3. RESULTS AND DISCUSSION

3.1. Willow

Average values of specific activity of two soils at Savichy fields and content of exchangeable forms of ^{137}Cs and ^{90}Sr as well as content of cations are presented in Table 3.

Table 3. Content of cations and radionuclides in soils of Savichy test fields

Soil type	Cations, mg-eq/100g			Exchangeable forms, Bq/kg		Total radionuclide content, Bq/kg	
	K	Ca	Mg	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr
Sandy (field #1)	0.0388 (±0.0075)	0.890 (±0.101)	0.02	108.25 (±3.40)	399.5 (±5.5)	4030 (±242)	472.5 (±10.7)
Peaty (field #2)	0.155 (±0.062)	11.13 (±0.22)	1.18 (±0.16)	2985 (±359)	10550 (±790)	139350 (±26510)	14810 (±1010)

In our example, peaty soil (field #2) contained approx. 12 times as much of exchangeable forms of Ca and 4 times as much of that of K compared to sandy soil (field #1). Under this condition, for the peaty soil, equilibrium between the nature analogues (K – Cs, Ca – Sr) is shifted towards calcium and potassium. As a result, a ratio of exchangeable forms of radionuclides to their total specific activity is lower for a peaty soil than for a sandy one (for 137Cs, see Figure 1).

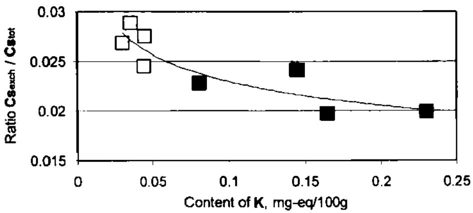

Figure 1: Relation between concentration of ^{134}Cs exchangeable forms and K content for Savichy fields. Blank boxes refer to sandy soil

Since exchangeable forms of the given radionuclides are biologically easy assimilable, the noted difference results in the fact that extraction of ^{137}Cs and ^{90}Sr from sandy soil is more intensive (Figure 2). The data obtained also confirm that there is no significant difference of accumulation coefficient between different clones of willow. In the given paper, the accumulation coefficient is defined as a ratio of radionuclide specific activity in willow tissue to that in soil.

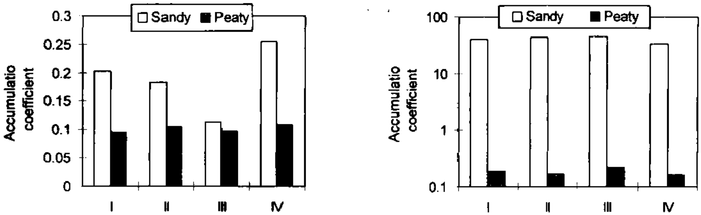

Figure 2: Accumulation coefficient of ^{137}Cs (left picture) and ^{90}Sr (right picture) for willow growing on different plots in Savichy

The results of specific activity measurements carried out at Masany field (Table 4) allows to evaluate the accumulation coefficient both for willow stem and leaves. In all plots of Masany field, the ^{137}Cs accumulation coefficient for leaves compared to stem is about sixfold higher. As to ^{90}Sr, the accumulation coefficient for leaves is by a factor of 1.5-3.5 higher compared to that for stem. Like in Savichy, we did not reveal any significant difference of accumulation coefficient between different clones of willow.

Table 4. Content of radionuclides in soils and willow of Masany test fields

Soil type	^{137}Cs specific activity, Bq/kg			^{90}Sr specific activity, Bq/kg		
	Soil	Leaves	Stem	Soil	Leaves	Stem
Peaty (field #4)	30920±3950	3566±808	598.8±215.2.	28780±12120	5856±1400	1998±784

3.2. Rape

The results of measurements of materials sampled from the rape test plantation are presented in Table 5. The same pattern of correlation between the content of exchangeable forms of radionuclides in soil and the soil cation exchange capacity is revealed (for ^{90}Sr, see Figure 3). From Figure 4 one can see that the accumulation coefficient of ^{90}Sr is about two orders of magnitude higher than that of ^{137}Cs. In different parts of *Brassica juncea*, ^{137}Cs accumulation coefficient conforms to the following scale: stems/leaves/flowers = 1/1.3/2.5. As to ^{90}Sr accumulation, this order is 1/8.5/3.

Table 5. Content of radionuclides in soils and plants of the rape test fields

Test plot #	^{137}Cs specific activity, Bq/kg			^{90}Sr specific activity, Bq/kg		
	Soil (total)	Soil (exchangeable)	Rape plant	Soil (total)	Soil (exchangeable)	Rape plant
1	1530	186	464.0	164	127	5278
3	1930	217	542.2	152	120	5296
5	1515	182	374.0	151	114	5756
7	1200	126	285.1	164	130	5692
9	1425	160	378.0	198	145	4504

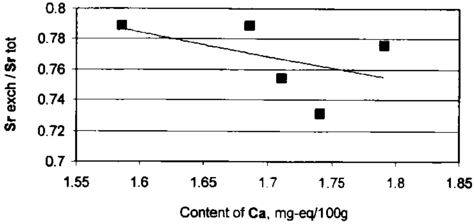

Figure 3: Relation between concentration of ^{90}Sr exchangeable forms and Ca content in rape test plots

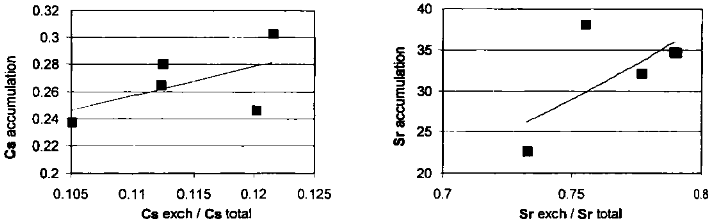

Figure 4: Accumulation coefficient of ^{137}Cs (left picture) and ^{90}Sr (right picture) for rape plant

4. CONCLUSION

The investigated correlation between the soil properties and the rate of extraction of radionuclides by the identified crops provides more data for sound decision on applicability of phytoremediation to radioactively contaminated area. For example, in view of relatively high ^{137}Cs transfer factor for willow wood, ranging from 0.2 10^{-3} through 1.5 10^{-3} m^2/kg, willow chips may not be applicable as a fuel if wood is extracted from the area of contamination of higher than 740 Bq/m^2. Otherwise, it will lead to excess of exemption limit established for wood fuel and to additional threat to population health.

Acknowledgment

The study was supported by RECOVER Project of the European Commission (DG-XII) and Initiatives for Proliferation Prevention Program of the US Department of Energy. The authors acknowledge a valuable assistance by Dr. Arnold A. Gvozdev, Institute of Radio-Ecological Problems, Belarus.

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