

# Extraction of $^{137}\text{Cs}$ by alcohol-water solvents from plants containing cardiac glycosides

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## Abstract

As a result of nuclear power plant accidents, large areas receive radioactive inputs of  $^{137}\text{Cs}$ . This cesium accumulates in herbs growing in such territories. The problem is whether the herbs contaminated by radio-cesium may be used as a raw material for medicine. The answer depends on the amount of  $^{137}\text{Cs}$  transferred from the contaminated raw material to the medicine. We have presented new results of the transfer of  $^{137}\text{Cs}$  from contaminated *Digitalis grandiflora* Mill. and *Convallaria majalis* L. to medicine. We found that the extraction of  $^{137}\text{Cs}$  depends strongly on the hydrophilicity of the solvent. For example 96.5%(vol.) ethyl alcohol extracts less  $^{137}\text{Cs}$  (11.6 %) than 40%(vol.) ethyl alcohol or *pure water* (66.2 %). The solubility of the cardiac glycosides is inverse to the solubility of cesium, which may be of use in the technological processes for manufacturing ecologically pure herbal medicine.

**Key words:** Herbal raw material; Herbal medicine;  $^{137}\text{Cs}$  contamination;  $^{137}\text{Cs}$  transfer.

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## 1 Introduction

At the present time, herbal medicines containing cardiac glycosides are widely used in the medical treatment of heart disease as the equivalent synthetic analogues are not available yet. The *Convallaria* and *Digitalis* species are used as raw material for the herbal cardiac medicine.

As a result of nuclear power plant accidents, large areas receive radioactive inputs of  $^{137}\text{Cs}$ . This cesium accumulates in herbs growing in such territories. The problem is whether the herbs contaminated by radiocesium may be used as a raw material for medicine. The answer depends on the amount of  $^{137}\text{Cs}$  transferred from the contaminated raw material to the medicine.

After the Chernobyl catastrophe a large area, growing many species of medicinal plants (such as *Convallaria majalis* L., *Rhamnus catartica* L., *Acorus calamus* L., *Vaccinium vitis-idaea* L., *Vaccinium myrtillus* L. etc.) was contaminated with a high concentration of radionuclides. The main contaminant (more than 90% of the overall radioactivity) of the Ukrainian Polessie is  $^{137}\text{Cs}$  [1]. The majority (85 to 97%) of  $^{137}\text{Cs}$  is located in the soil layer at a depth of 0 to 10 cm, where the roots of the medicinal herbs take up the  $^{137}\text{Cs}$  [2].

Various solvents are used in the pharmaceutical industry to prepare the herbal medicinal products. The properties of these solvents define a quantitative and qualitative structure of substances that can be extracted. The literature gives some often contradictory information on the transfer of  $^{137}\text{Cs}$  from medicinal plant raw material to water and alcohol. Several studies [3, 4, 5, 6, 7] have shown that the amount of radionuclide transferred from the soil to the raw material varied in wide range (from 10 to 250 fold). This difference was mainly determined by specific features of the plants and depended strongly on the type of soil and climatic conditions which occurred during the vegetative period. The transfer of  $^{137}\text{Cs}$  from the plant raw material is 24-75% for the aqueous medicinal products and is 20-30% for the alcoholic ones [8, 9, 10, 11]. Grodzinski et al. [12] found that the specific activity of  $^{137}\text{Cs}$  in water extracts was three orders of magnitude less than in the initial plant raw material.

One can conclude, from the above publications that experiments have been mainly devoted to studying the transfer of  $^{137}\text{Cs}$  from plant raw material to water and some galenical preparations. Systematic studies of the dependency of radionuclide extraction efficiency on various types of solvents have not been done yet. Therefore the study of radionuclide transfer from soil to herbs and from herbs to the herbal medicinal products is very important.

In this paper we present our experimental results on the effect of solvent type (*pure water*, *40%(vol.)*, *70%(vol.)* and *96.5%(vol.) aqueous ethyl alcohol*) on  $^{137}\text{Cs}$  extraction efficiency.

## 2 Methods

## 2.1 Raw material

We studied the medicinal plant species (the herbs *Digitalis grandiflora* Mill, flowers and leaves of *Convallaria majalis* L.) containing cardiac glycosides. The raw material was taken from an experimental plot of the Povchansk forest area, Luginy district, Zhitomir region where the soil contamination by  $^{137}\text{Cs}$  ranged from 296 to 925  $\frac{\text{kBq}}{\text{m}^2}$  and the radioactivity of the tested plant raw material was 1.17 – 50.83  $\frac{\text{kBq}}{\text{kg}}$  (see Table 1). All raw plant material was rolled three times in preparation for the extraction process.

## 2.2 Tinctures and aqueous extracts

We used the following solvents: *pure water*, *40%(vol.)*, *70%(vol.)* and *96.5%(vol.) aqueous ethyl alcohol* without heating and vaporization of solvent. Samples (10.0 g aliquots) of raw material were put in glass extractors, filled up with the appropriate concentration of solvent and let stand for extraction. After 48 hours, the extracts were discharged to a flask and the extractors were filled up with a new portion of the solvent for further extraction. The operation was repeated after 24 hours and 48 hours to obtain the second and third extracts. Then the solvent residuals were removed using vacuum and added to the extracts. All extracts were combined to obtain the primary tinctures of 100 ml volume.

## 2.3 Decoctions

The solvent that we used was hot water. The 10.0 g samples of raw material were put in glass vessels, filled up with pure water and boiled for 15 minutes. After that these vessels were cooled and kept at room temperature for 10 minutes. The filtered extracts and the solvent residuals were then combined to give the decoctions of 100 ml volume.

## 2.4 Measurement of radioactivity

The  $\gamma$  activity of both the initial raw material and the extracts were measured during the experiment. The standards of the Health Ministry of Ukraine state that the specific activity of  $^{137}\text{Cs}$  in medicinal plants must be less than 600  $\frac{\text{Bq}}{\text{kg}}$  [13]. We needed to measure a small activity and commercial devices (like the *LP - 4900B*) have insufficient sensitivity, therefore we used the  $\gamma$  detector from the Institute for Single Crystals (Kharkov, Ukraine). The scintillator (*BGO*) was a cylinder with  $d=40\text{ mm}$  and  $h=40\text{ mm}$ . The detector was calibrated with  $^{60}\text{Co}$  ( $E_\gamma=1.173\text{ MeV}$ ,  $E_\gamma=1.332\text{ MeV}$ ) and  $^{137}\text{Cs}$  ( $E_\gamma=0.662\text{ MeV}$ ) sources from a standard calibration set before measurements. For the  $E_\gamma=0.662\text{ MeV}$  photon the energy resolution of the detector was determined to be 13.4%. The photopeak efficiency of photon registration for the point source was  $\varepsilon=0.42$ . The detector and samples were placed inside a 5 cm thick lead shield to decrease the background. The background at the 0.662 MeV peak region was about 0.2  $\frac{\text{count}}{\text{sec}}$ . The samples were measured directly inside Marinelli's vessel (100 ml

volume). The detector efficiency was  $31.5 \left[ \frac{Bq \times sec}{count} \right]$ . The activity of  $^{137}\text{Cs}$  in the samples was determined by formula  $A \left[ \frac{Bq}{kg} \right] = 31.5 \times N_{ph.p.}$ , where  $N_{ph.p.} \left[ \frac{1}{sec \times kg} \right]$  is the number of counts in the photopeak of the measured substance.

The data acquisition system was assembled in CAMAC standard. The  $\gamma$  spectrum parameters were calculated with the MINUIT program from the ROOT package [14].

All samples were measured over a wide range of photon energy (0.1 – 2.0 MeV). Only one peak at 0.662 MeV was detected which meant that  $^{137}\text{Cs}$  was present and therefore we only studied the transfer of  $^{137}\text{Cs}$ . The statistical error of the measurements did not exceed 5%. The absolute error was 15% and depended on the standard calibrated source of  $^{137}\text{Cs}$ .

### 3 Results and discussion

The results of the determination of  $^{137}\text{Cs}$  transfer from the herbs to alcohol-water and pure water extracts are presented in Table 1. From the table one can see that for the plants investigated, the  $^{137}\text{Cs}$  transfer does not depend on the type of raw material (within the error limits). The reason for certain scattering of the data for  $^{137}\text{Cs}$  transfer for 96.5%(vol.) alcohol can be due to saturation of solvent by cesium.

Studying the dependence of the  $^{137}\text{Cs}$  transfer on the solvent hydrophilicity shows that for 96.5%(vol.) alcohol the transfer of  $^{137}\text{Cs}$  to the extract is minimal, ranging from 4.6 to 17.6%. For 70%(vol.) alcohol the transfer reaches 45.3 to 66.5%. In our experiments the maximal amount of  $^{137}\text{Cs}$  was extracted by 40%(vol.) alcohol (62.8 to 83.2%) and pure water at room temperature (63.0 to 73.0%). Decoctions extracted radiocesium similar to 70%(vol.) alcohol (49.3 to 60.8%) but less than 40%(vol.) alcohol or pure water at room temperature. The reason is that in the process of heating some of the cesium chemically interacts with the raw material and can not be extracted.

As follows from our results the 96.5%(vol.) alcohol extracts less (about 6 times)  $^{137}\text{Cs}$  than 40%(vol.). From Table 2 one can see that solubility of the cardiac glycosides strongly depends on solvent type and the solubility tendency of the cardiac glycosides is inverse to cesium [15]. Methanol extracts the cardiac glycosides much more (a factor of 570 for *Digitoxin* and a factor of 18 for *Convallatoxin*) as compared to water. Thus the ratio of  $^{137}\text{Cs}$  to the cardiac glycosides extracted strongly depends on solvent type.

### 4 Conclusion

We have presented new results on the transfer of  $^{137}\text{Cs}$  from raw material to medicine. We have found that the extraction of  $^{137}\text{Cs}$  from *Digitalis grandiflora Mill.* and *Convallaria majalis L.* containing cardiac glycosides strongly depends on solvent hydrophilicity and where 96.5%(vol.) alcohol extracts less  $^{137}\text{Cs}$  (about 6 times) than 40%(vol.) or *pure water*. The solubility tendency of

the cardiac glycosides is inverse to that of cesium and this fact can be of use in the technological processes for manufacturing ecologically pure herbal medicine.

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Table 1  
<sup>137</sup>Cs transfer from medicinal plant raw material to alcohol and water extracts

Herbs	Specific <sup>137</sup> Cs activity in soil, $kBq \times m^{-2}$	Specific <sup>137</sup> Cs activity in raw material, $kBq \times kg^{-1}$	<sup>137</sup> Cs transfer to tinctures, %			<sup>137</sup> Cs transfer to water, % extracts at room temperature	
			96.5 <sup>0</sup>	70 <sup>0</sup>	40 <sup>0</sup>	decoctions	
<i>Herb of Digitalis No1</i>	925	2.66	11.6	46.6	66.2	56.4	66.2
<i>Herb of Digitalis No2</i>	814	1.48	17.6	45.3	62.8	49.3	68.9
Average			14.6	46.0	64.5	52.9	67.6
<i>Leaves of Convallaria No1</i>	296	1.49	10.7	63.1	67.1	60.0	74.0
<i>Leaves of Convallaria No2</i>	777	50.83	6.3	59.7	63.9	59.1	73.0
Average			8.5	61.4	65.5	59.6	73.5
<i>Flowers of Convallaria No1</i>	740	11.73	4.6	58.9	66.0	60.8	69.1
<i>Flowers of Convallaria No2</i>	296	2.81	10.0	66.5	69.4	67.3	71.5
<i>Flowers of Convallaria No3</i>	407	3.64	14.0	52.7	83.2	59.6	63.0
Average			9.5	59.4	72.9	62.6	67.8

Table 2  
Solubility of some cardiac glycosides

Glycoside	Herb	Part of solvent needed to dissolve one part of glycoside	
		water	methanol
Digitoxin	<i>Digitalis</i>	40 000	70
Convallatoxin	<i>Convallaria</i>	1000	56