LOW LEVEL MEASUREMENTS OF RADIOACTIVE RESIDUALS AS SPIN-OFFS FROM HIGH ENERGY AND PARTICLE PHYSICS TO THE MEDICINE AND NUTRITION QUALITY CONTROL SERVICE

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ABSTRACT

As a result of nuclear power plant accidents, large areas receive radioactive inputs of ^{137}Cs. Some quantity of radioactive elements can penetrate into the soil and be accumulated by herbs. In fact, such radioactive contamination can further appear in herbal medicines and nutrition. Even modest concentration of such radioactive contamination, if presented in medical products and nutrition, can cause severe human diseases. To ensure high quality of medical products, the Quality Control Service should be capable to measure low level radiation in products. According to the standards of Health Ministry of Ukraine, the ^{137}Cs specific activity of medicinal plants must be less than 600 *Bq/kg* (~272 *Bq/lb*). The studies of the ^{137}Cs transfer from contaminated medical raw materials such as Digitalis grandiflora and Convallaria majalis to medicines were performed by Experimental Nuclear Physics, Solid State Physics, and Medical Chemistry groups. Studies showed that the extraction of ^{137}Cs depends strongly on the hydrophilicity of the solvent. For example, 96.5% ethyl alcohol extracts less ^{137}Cs (11.6%) than 40% ethyl alcohol in pure water (66.2%). The solubility of the cardiac glycosides is inverse to that of cesium, which may be used in the technological processes for manufacturing ecologically pure herbal medicines and nutrition. Such studies are an excellent example of the spin-offs form Nuclear Physics to Medicine.

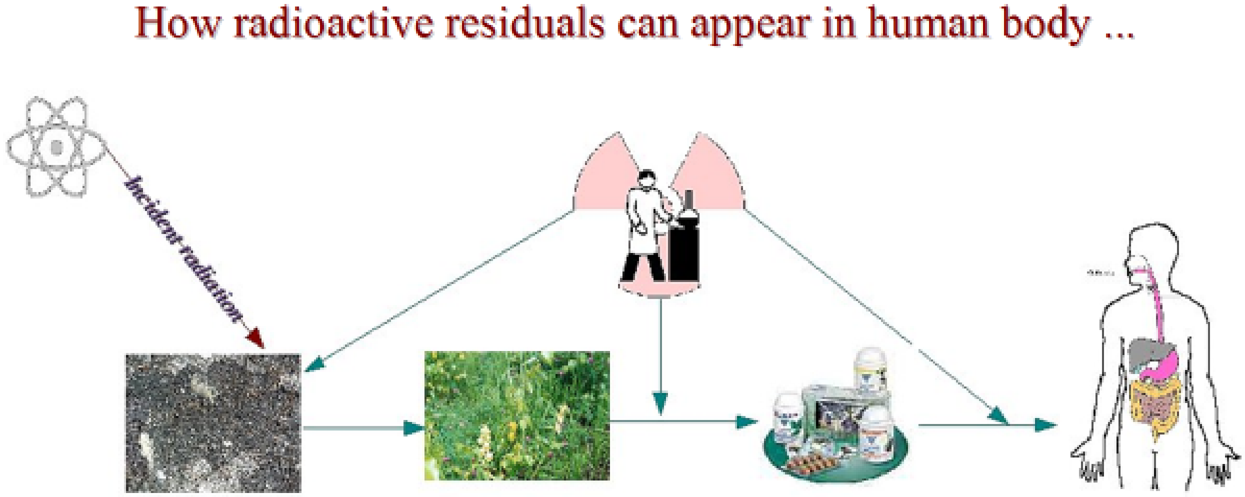
Figure 1. Radiation hotspots resulting from the Chernobyl nuclear power plant accident.

INTRODUCTION

As a result of the Chernobyl (Ukraine) nuclear power plant accident, the large area of Polessie received radioactive inputs {Figure 1}.

The main contaminant (more then 90% of the overall radioactivity) of the Ukrainian Polessie is ^{137}Cs (Buzun et al., 1999). The majority (85 to 97%) of ^{137}Cs is located in the top soil layer (10cm or 4in) where the roots of the medicinal herbs take up the ^{137}Cs (Pushkarev et al., 1997). If such herbs were used for medicine and

Figure 2.



Pictures: http://www.geforce.dk/archive/window/ (radiation); file:///home/sveta/ttt/ground.htm (ground); http://www.mig19.schel.ac.ru/pages/herbarium/photos/fnaper.jpg (meadow); http://www.kzb.ru/clauses/krestianka.html (herb medicine); http://cyro.cs-territories.com/gcse_biology/biohumandigq.jpg (human); http://www2.fpm.wisc.edu/safety/Radiation/trainman.htm (radiation safety)

For years Polessie has been traditionally used for industrial harvesting of medicinal plants and now the problem is whether or not the herbs from contaminated areas may be still used for medicines. An answer depends on the amount of ^{137}Cs transfered from the contaminated raw material to the medicine. Thus special studies should be done to determine the optimal extraction technique which can even suppress occurring ^{137}Cs in medicines.

In the pharmaceutical industry various solvents are used 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}Cs from medicinal plant raw materials to water and alcohol. Several studies (cf. Orlov et al., 1996; Krasnov et al., 1996; Orlov et al., 1997; Sanarov et al., 1998; Grischenko et al., 1990) have shown that the amount of radionuclide transferred from the soil to the raw materials varied in wide range (results differ by factor 250!). This difference was mainly determined by specific features of the plants and it strongly depended on the type of soil and climatic conditions which occurred during the vegetative period. The transfer of ^{137}Cs from the plant raw material is 24-75% for the aqueous medicinal products and is 20-30% for the alcoholic ones (cf. Antonova and Seditzkaia, 1989; Prokofiev et al., 1992; Prokofiev et al., 1993; Dmitriev et al., 1991). Grodzinskii found that the specific activity of ^{137}Cs in water extracts was three orders of magnitude less than in the initial plant raw material (Grodzinskii et al., 1991).

From the above publications one can conclude that experiments have been mainly devoted to studying the transfer of ^{137}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.

nutrition, the ^{137}Cs could occur in human body and can cause severe disease {Figure 2}.

METHODS

Preparation of raw material. The following species, containing cardiac glycosieds, have been studied: the herbs of Foxglove (Digitalis grandiflora Mill) {Figure 3}, flowers and leaves of Lily-of-the-valley (Convallaria majalis L.) {Figure 4}. The raw materials were taken from an experimental plot of the Povchansk forest area (Luginy district, Zhitomir region, Ukraine) where the soil contamination by ^{137}Cs ranged 296 - 925*kBq/m*^{2} and the radioactivity of the tested raw plant material was 1.48 - 50.83 *kBq/kg* (see Table 1). Before extraction, harvested raw material was dried and passed through the roller press three times.

Figure 4. Lily-of-the-valley (*Convallaria majalis*) http://sad.zeleno.ru/enc.php?a=out

Figure 3. Foxglove (*Digitalis grandiflora*) http://sad.zeleno.ru/enc.php?a=out

Preparation of tinctures and aqueous extracts. The following solvents, without heating and vaporization, were used: pure water, 40%(vol.), 70%(vol.) and 96.5%(vol.) aqueous ethyl alcohol. Samples (10.0 g aliquots) of raw material were put in glass extractors (Figure 5), 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.

Radioactive residuals could transfer from herbs to medicine products

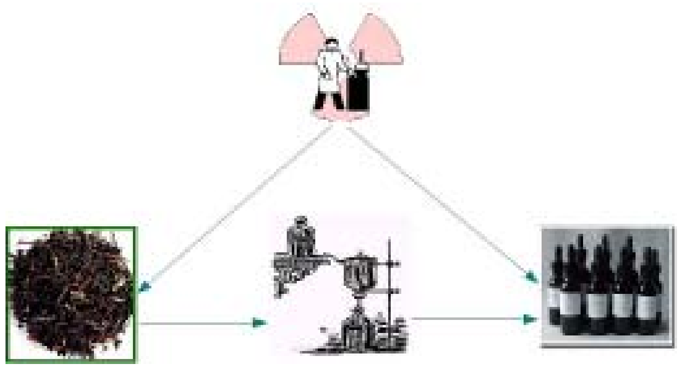


Figure 5. http://medic.oke.ru/people/nature/1_-0065-1.jpg (raw material); http://www.ibiblio.org/herbmed/eclectic/journals/pics/ajp1883-percolation.jpg (percolation); http://www.kerrysherbals.com/graphics/prod_tinctures.jpg (tinctures).

Preparation of decoctions (herbal tea). The 10.0 g samples of raw material were put in glass vessels, filled up with pure water and boiled for 15 minutes. Afterwards these vessels were cooled and kept at room temperature for 10 minutes. 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.

Measurement of radioactivity. The ã-activity of both the initial raw material and the extract were measured during the experiments. The standards of the Health Ministry of Ukraine state that the specific activity of ^{137}Cs in medicinal plants must be less than 600*Bq/kg*. Small activity levels should be measured, however, commercial devices available in Ukraine (LP 4900B and similar to that) have insufficient sensitivity. To overcome such a problem, unique hardware and software was designed within the interdisciplinary collaboration between Experimental Nuclear and Particle Physics, Solid State Physics and Medical Chemistry groups. The ã-detector was manufactured by the Institute for Single Crystals (Kharkov, Ukraine). The scintillator (BGO) was a cylinder with d=40*mm* and h=40*mm*. Before measurements the ã-detector was calibrated using ^{60}Co (Eã=1.173*MeV*, Eã=1.332*MeV*) and ^{137}Cs (Eã=0.662*MeV*) sources from a standard calibration set. For the Eã= 0.662*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 å=0.42. The detector and samples were placed inside the 5 cm thick lead shield to decrease the background. The background at the 0.662*MeV* peak region was about 0.2[*counts/sec*]. The samples were located inside Marinelli's vessel (100*ml* volume) where measurements were done. The detector efficiency was 31.5[*Bq*sec/count*]. The activity of ^{137}Cs in the samples was determined by formula:

$$A\ [Bq/kg] = 31.5 \cdot N_{ph.p.},$$

where $N_{ph.p}$ [$sec^{-1}*kg^{-1}$] is the number of counts in the photopeak of the measured substance.

The data acquisition system was assembled in the crate CAMAC standard. The ã-spectrum parameters were calculated using the MINUIT program from the ROOT package (cf. Brun and Rademakers, 1997). 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}Cs was present and therefore only the transfer of ^{137}Cs was studied. The statistical error of the measurements did not exceed 5%. The absolute error was 15% and depended on the standard calibrated source of ^{137}Cs.

RESULTS AND DISCUSSIONS

The results of the determination of ^{137}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 studied herbs, the ^{137}Cs transfer does not depend on the type of raw material (within the error limits) {see Figure 6}. The reason for certain divergence of data for ^{137}Cs transfer for 96.5%(vol.) alcohol could be due to saturation of solvent by cesium.

Studying the dependence of the ^{137}Cs transfer on the solvent hydrophilicity {Figure 7} showed that for 96.5%(vol.) alcohol the transfer of ^{137}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}Cs was extracted by 40%(vol.) alcohol (62.8 to 83.2%) and pure water at room temperature (63.0 to 74.0%). Decoctions extracted radiocesium (49.3 to 60.8%) similar to 70%(vol.) alcohol 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 ^{137}Cs less (about 6 times) than 40%(vol.) ones. From Table 2 one can see that solubility of the cardiac glycosides strongly depends on a solvent type and is inverse to the solubility of cesium (cf. Baumgarten, 1969). Methanol extracts the cardiac glycosides much more (a factor of 570 for Digitoxin and a factor of 18 for Convallatoxin) as compare to water. Thus the ratio of ^{137}Cs to the cardiac glycosides extracted strongly depends on

^{137}Cs transfer from medicinal plant raw materials to alcohol and water extracts

Table 1

Herbs	Specific ^{137}Cs activity in soil, *kBq/m²*	Specific ^{137}Cs activity in raw materials, *kBq/kg*	^{137}Cs transfer to tinctures, % (*vol*)			^{137}Cs transfer to water, %(*vol*)	
			96.5	70	40	decoctions	extracts at room temperature
Herb of Digitalis No1	925	2.66	11.6	46.6	66.2	56.4	66.2
Herb of Digitalis No2	814	1.48	17.6	45.3	62.8	49.3	68.9
Average			*14.6*	*46.0*	*64.5*	*52.9*	*67.6*
Leaves of Convallaria No1	296	1.49	10.7	63.1	67.1	60,0	74.0
Leaves of Convallaria No2	777	50.83	6.3	59.7	63.9	59.1	73.0
Average			*8.5*	*61.4*	*65.5*	*59.6*	*73.5*
Flowers of Convallaria No1	740	11.73	4.6	58.9	66.0	60.8	69.1
Flowers of Convallaria No2	296	2.81	10.0	66.5	69.4	67.3	71.5
Flowers of Convallaria No3	407	3.64	14.0	52.7	83.2	59.6	63.0
Average			*9.5*	*59.4*	*72.9*	*62.6*	*67.8*

^{137}Cs transfer from different part of plant to alcohol and water extracts

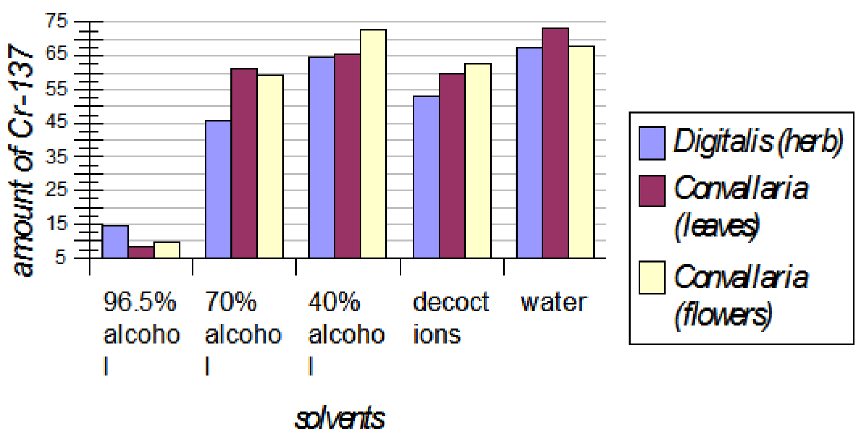
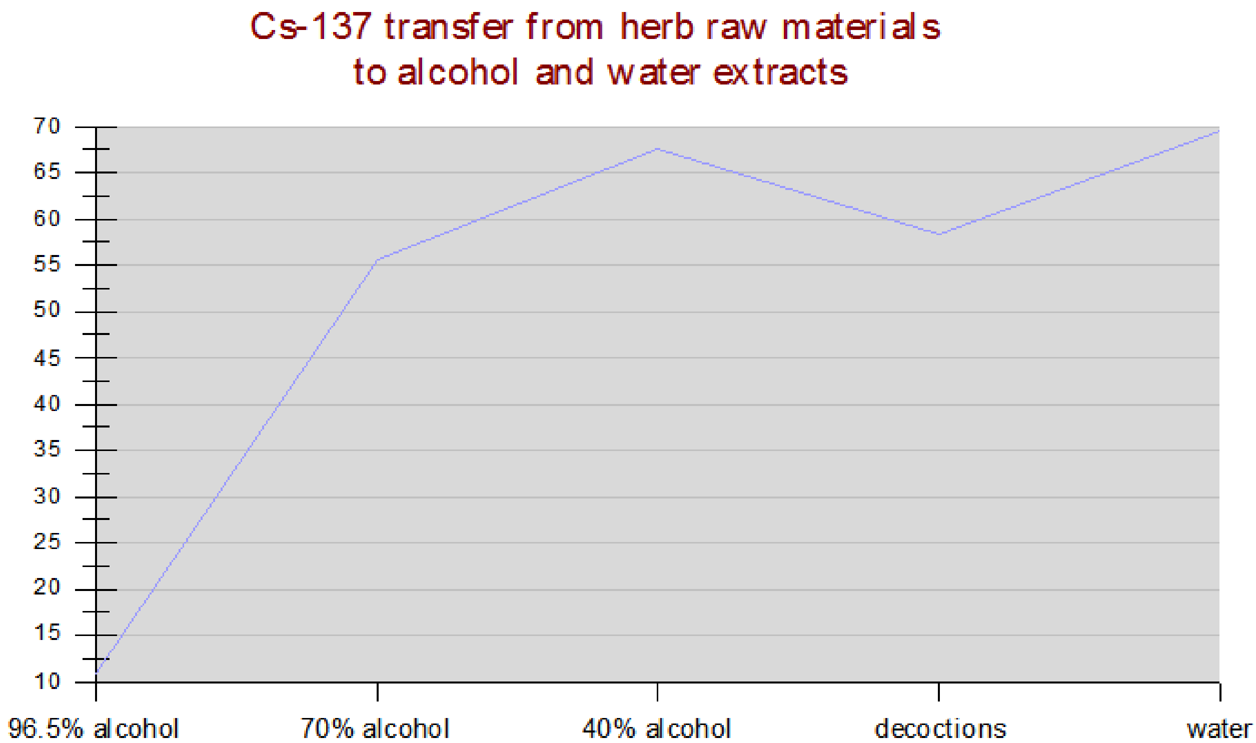


Figure 6.



Solubility of some cardiac glycosides

Table 2.	Glycoside	Herb	Part of solvent needed to dissolve one part of glycoside	
			water	methanol
	Digitoxin	Digitalis	40 000	70
	Convallatoxin	Convallaria	1000	56

solvent type.

CONCLUSIONS

The new results of transferring ^{137}Cs from raw materials to medicines have been presented. It was found that the extraction of ^{137}Cs from Digitalis grandiflora Mill. and Convallaria majalis L. containing cardiac glycosides strongly depends on solvent hydrophilicity where 96.5%(vol.) alcohol extracts about 6 times less ^{137}Cs than 40%(vol.) alcohol 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 medicines.

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