



The characterization and removal of Chernobyl debris in garden soils

Andersson, Kasper Grann

Publication date:
1991

Document Version
Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

Citation (APA):
Andersson, K. G. (1991). *The characterization and removal of Chernobyl debris in garden soils*. Risø National Laboratory. Risø-M No. 2912

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

DK9100033

RISØ

Risø-M-2912

The Characterization and Removal of Chernobyl Debris in Garden Soils

Kasper G. Andersson

**Risø National Laboratory, DK-4000 Roskilde, Denmark
January 1991**

The Characterization and Removal of Chernobyl Debris in Garden Soils

Riso-M-2912

Kasper G. Andersson

*Risø National Laboratory, DK-4000 Roskilde, Denmark
January 1991*

Abstract

Severe nuclear accidents such as the one in Chernobyl in 1986 may give unacceptably high external radiation levels, which even in the late phase may make a resettlement of an evacuated population impossible unless action is taken to decrease the exposure.

As the urban land areas to be reclaimed may be very large the cost of the dose reducing countermeasure to be used may be an important factor. In the Chernobyl debris the most important radionuclides concerning the long term external radiation were found to be Cs-137, Cs-134, and Ru-106. Therefore, the aim of this work is to investigate the behaviour of these radionuclides in garden soils, and on this background to examine cost-effective methods by which a reduction of the dose from such areas to people living in urban or sub-urban environments can be achieved.

The fixation of the radioactive cations in soil was investigated by means of soil profile sampling, soil texture analysis, and speciation experiments. It was found that most of the Chernobyl fallout caesium was extremely firmly fixed. Much of the ruthenium was more loosely bound, to organic material.

The cost-effectiveness of some dose reducing countermeasures was examined on the background of small scale tests. Here it was found that about 95 % of the activity could be removed with peelable fixatives based on PVA or lignin.

ISBN 87-550-1699-5

ISSN 0418-6435

Grafisk Service, Riss 1991.

TABLE OF CONTENTS

1. INTRODUCTION..... 5

2. PRESENTATION OF THE SURFACE TO BE DECONTAMINATED..... 6

2.1. MECHANISMS OF CATION FIXATION IN SOILS..... 6

2.2. EXPERIMENTAL INVESTIGATIONS OF SOIL PROFILES..... 8

2.3. SPECIATION EXPERIMENTS..... 12

3. THE EFFICIENCY OF RECLAMATION PROCEDURES FOR GARDENS..... 19

3.1. PVA COATING..... 19

3.2. LIGNOSOL BD COATING..... 21

3.3. OTHER COATINGS..... 22

4. SUMMARY AND CONCLUSIONS..... 23

5. ACKNOWLEDGMENTS..... 23

6. REFERENCES..... 24

1. Introduction

In the years following the Chernobyl accident an extensive research programme has resulted in the identification of the most important pathways of external radiation, and has led to the identification of some of the mechanisms in the fixation of radionuclides in the urban environment.

It was revealed (Roed et al. 1990) that generally the largest contribution to the dose rate came from the open areas such as gardens and parks, and thereafter the roads, pavements and walkways.

The major part of the dose rate in all investigated urban environments came from the deposition on open (grassed) areas.

Therefore it is important to investigate means by which the dose rate from such surfaces can be reduced.

To investigate the applicability of different methods for dose reduction it is necessary to consider the type of environment in which it is applied.

In the past, research in this field has focused on the rural areas, in which for instance ploughing is a possibility for reducing dose.

However, due to the topography, a procedure of this type would be impossible to perform on most grassed/open areas in the urban environment, and other methods have been recommended for this purpose.

Mainly on the background of pre-Chernobyl research the IAEA (IAEA, 1987) described some of the main problems of the suggested countermeasure. Following a grass cutting and removal of vegetables, bushes, flowers, etc., the recommendation was simply to remove the uppermost 5-10 cm of soil. From a garden area of 200 m² this would generate 10-20 cubic meters of soil waste, in other words several truck loads.

A removal of the arable top soil layer seemed to be the only practicable way to eliminate the contamination. However, if the garden was dug and the contamination was buried at the depth of one spit, the resultant shielding effect is (Gjørup et al. 1982) a reduction of the dose by a factor of 6.

This latter procedure could be performed relatively easily and would not require the use of sophisticated tools. However, the activity would still be present in the soil.

If the thickness of the removed top soil layer could be reduced substantially, it would be a great advantage because the land areas would still be fully arable, and the amount of radioactive waste

generated would be smaller. As the soil surface is not plane enough to allow this to be accomplished by scraping off a thin layer, other methods needs to be considered.

An alternative means for removing the surface layer is the utilization of strippable coatings. A number of coatings or "fixatives" have been investigated for different purposes.

The term "fixative" as such refers simply to a material which may be used to bind particles to a surface. The purpose of most previous applications of fixatives has been to prevent a resuspension which might occur either by wind or due to other effects. In these cases the fixation will greatly reduce the inhalation hazard.

Apart from these possibilities fixatives could also be used in connection with decontamination procedures. Here, a fixation of the radioactive substances to the removed surface could bring about an increasing efficiency of the procedure.

Furthermore, in a soil the thickness of the removed surface layer would be determined by the viscosity of an applied liquid fixative which mixes with the soil particles in the thin upper layer. Thereby, the dried fixative would enclose the top layer in which the major part of the activity will be found right after deposition. The activity would therefore be embedded in a relatively thin film which could be removed.

To optimize the effect of any reclamation procedure a knowledge of the nature of the contamination will be needed.

2. Presentation of the Surface to be Decontaminated

2.1. Mechanisms of Cation Fixation in Soils

Contaminated areas of soil are difficult to reclaim by leaching because of the great potentials of the soil for cation exchange. This was demonstrated in an experiment (Nishita & Essington 1966) in which a range of different freshly contaminated soils were leached with various solutions. The obtained downward migration was small, although a fraction of the multivalent contaminants, such as ruthenium-106, seemed to be leachable to some extent.

Two different types of soil components are responsible for this fixation. The organic and the inorganic. Which of the effects is

the dominant depends on the nature of the contaminant ion and on the soil characteristics. Cations which do not have a tendency to be hydrated when located in the clay interlayer space can be captured very efficiently by the clay structure.

Caesium is such an ion, and is therefore strongly bound by certain minerals, such as illite. Ruthenium, another important ion in the debris from Chernobyl, is not trapped as efficiently by the clay.

However, the effect of fixation by sequestering organic ligands is more marked for multivalent ions like for instance ruthenium. Actually, the total cation exchange capacity of the humic part of the soil may be as much as a hundred times that of the clay part (Schnitzer & Khan 1978). The organic cation exchange capacity decreases with the soil pH (Brady 1970).

It may be difficult to determine, whether cations are kept from migrating by the clay fraction or the organic, as the humic soil substances consist of a complexity of organic material bound to the surfaces of a mixture of clay minerals by inter-ion coulomb forces and van der Waals attractions between molecules. Actually, the fraction of organic matter combined with inorganic matter is only less than 50 % in extremely sandy soils (Schnitzer & Kahn 1978).

There are two possible types of fixation in soil organic material. One is by simple cationic exchange with more or less loosely bound ions. The other is the chelating effect, which is more stable, and not so easily disturbed by waterflow or microbial attacks.

A chelate is simply a complex which is formed when an ion and an organic molecule are bound to each other by more than one bond, thereby together forming a cyclic structure. Although chelate formations can have different degrees of stability (Allison 1973), they are usually very stable. The stability increases with the number of atoms which form a ring with the captured cation and with the number of rings participating (Chaberek & Martell 1980).

It has been reported (Duff et al. 1963) that the activity of certain acid-producing bacteria may cause a considerable solubilization and subsequent chelation of cations from various humic substances, and in certain cases (2-ketogluconic acid) even from minerals, which are normally considered insoluble. It is also emphasized that smaller chelating agents such as fulvic acid may have a very important role concerning the downward migration of cations in a soil profile.

2.2. Experimental Investigations of Soil Profiles

In order to investigate the nature and degree of fixation of Chernobyl fallout in heavily contaminated soils, soil cores were sampled from the Chernobyl area, four and a half years after the contamination took place. The activity profiles of the radionuclides were as shown in fig. 1. The examined soil was a typical Chernozem, which is characterized by a very dark brown humic surface horizon which grades below into a lighter chestnut colored soil. Chernozem soils are particularly rich in organic content and in fine particles (less than 0.01 mm).

It has been stated (Bunzl et al. 1989) that on the grounds of speciation experiments, Chernobyl radiocaesium appears to be more labile than bomb fallout caesium. This could be interpreted as a result of the caesium being of a different form at the time of deposition, but may also simply reflect the time-dependent "locking up" of the caesium in the soil structure. It is well known that the strength of fixation increases with time (Squire & Middleton 1966).

Figure 2 shows the time dependence of two Cs-137 profiles of different soils from the same area in Gävle in Sweden. Each profile represents a 7th order polynomial smoothing of 5 individually sampled profiles, consisting of approximately 1 cm thick slices.

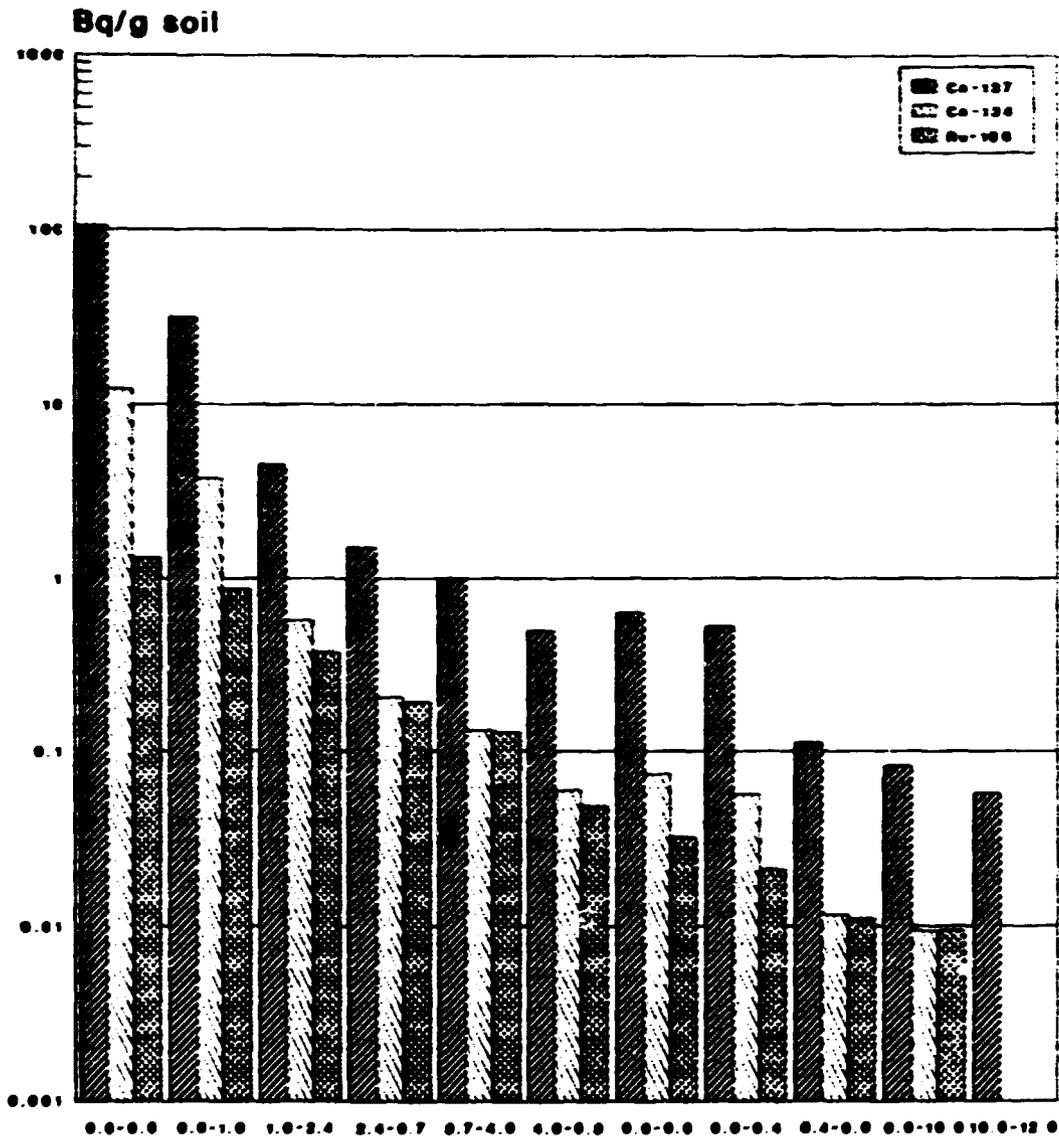
The characteristics of the two soils are given in table 1. The profiles are remarkably different. In soil A 90 % of the activity lies in the upper 1 cm. The maximum concentration in soil B is found approximately 2 cm down in the soil. As the fixation of caesium in soil is usually related to the presence of clay minerals, one might expect soil A to have the highest clay content. However, as can be seen from table 1, this is not the case. But the humic content of soil A is unusually high. As can be seen, the pH of soil A is also higher than that of soil B. Therefore, the total number of cation exchange sites may be much larger. This might be an explanation for the difference.

The profiles were measured 2 and 4 years after deposition, and whereas soil B shows no sign of downward migration of radionuclides, a small tendency can be seen in the case of soil A. This could be due to the degradeability of the organic traps (In certain soils the amount of caesium bound to insoluble organic chelated complexes can be relatively large (Route 1990), (Livens & Baxter 1988)).

Figure 1.

Cs-137 Cs-134 Ru-106

Soil Profiles from Chernobyl



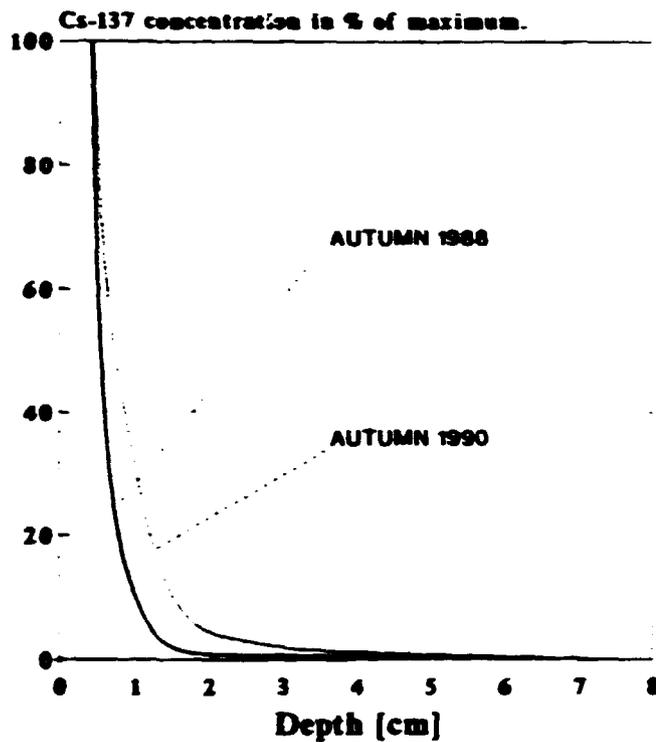
Bq/g soil

Cs-137	105.00	21.00	6.00	1.01	0.970	0.400	0.400	0.30	0.112	0.050	0.050
Cs-134	10.00	2.70	0.970	0.200	0.100	0.05	0.050	0.050	0.072	0.050	
Ru-106	1.01	0.97	0.200	0.100	0.100	0.050	0.050	0.050	0.051	0.050	

Figure 2.

Chernobyl Cs-137 Levels in Gävle Soil

SOIL A



SOIL B

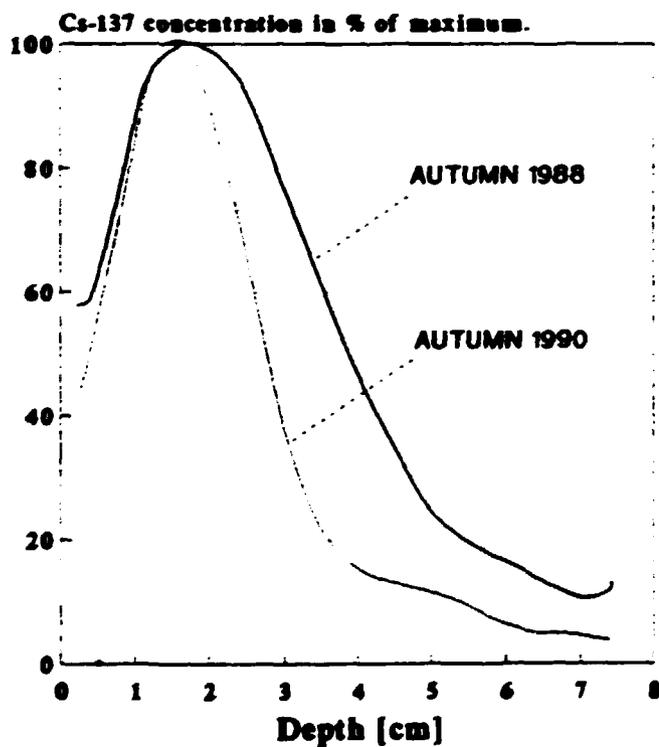


Table 1. Characteristics of two soils in Gävle in Sweden. Texture analysis performed by Nedeselskabet in Viborg, Denmark after the "hydrometer" method.

	Humus organic	Clays < 2µm	Silts 2- 20µm	Sand 1 20-200µm	Sand 2 0.2-2mm	pH
Soil A :						
Top 2cm	10.0%	6.8%	12.7%	45.2%	25.3%	6.5
3-5 cm	5.4%	6.6%	14.0%	50.9%	23.1%	
Soil B :						
Top 2cm	2.4%	13.1%	19.2%	49.7%	15.6%	4.7
3-5 cm	1.9%	13.0%	19.1%	51.3%	14.7%	

On the grounds of the texture analysis both soils are characterized as sandy loams.

The reason for the difference could also be sought in the deposition phase. A relatively rapid vertical transport of radionuclides in the soils may occur with a heavy rainshower, due to the hydrodynamics of the soil water flux. Here factors such as cracks and fissures, relative to incomplete wetting may have resulted in a different deposition pattern. Incomplete wetting, however, is not a very credible explanation, as the geographical distance between the two sampling areas was so small that the amount of rainfall can be assumed to be identical.

Anyway, it can be seen that there has been very little, if any, vertical migration in the period between the two samplings. The shorter tail on profile B in 1990 compared to 1988 may be the result of an increased transfer rate through a visibly much sandier layer located approximately 5 cm down in the profile.

From the Chernobyl area profiles, where ruthenium levels were still measurable, it can be seen that the ruthenium, which is much more widely associated with organic matter, has migrated much deeper into the soil than the caesium isotopes. About 72 % of the caesium is still found in the top 8 mm four and a half years after deposition, but only 44 % of the ruthenium is found in this layer. This, evidently, is due to the generally weaker fixation provided by ruthenium-organic bonds.

2.3. Speciation Experiments

In order to investigate the strength of fixation of caesium and ruthenium in the Chernobyl soil samples, speciation experiments were carried out by a modified version of the method introduced by Tessier et al. (1979).

This method of sequential "selective" extractions was altered by Salbu et al. (1989). The fractionation procedure introduced below is a slightly modified version of the one presented by Salbu.

The extractions were performed as follows:

1) First, the soil was dried at 50 °C and sieved so that only the <2mm fraction was included. Then a sample of approximately 2g was weighed and accordingly analysed for radionuclide content, using a lead shielded 15.6 % efficiency Ge(Li) detector. Hereafter the samples were subjected to extraction as described in the following sequence.

2) Fraction 1 : The first extraction was performed in order to determine the very loosely bound (readily exchangeable) fraction of the contaminants. For this purpose 20 ml of the inert electrolyte 1 M $\text{CH}_3\text{COONH}_4$ (NH_4Ac) (at pH 7) was applied and the suspension stirred rapidly for 2 hours at room temperature. Hereafter the liquid was separated from the solid phase by centrifugation at 3300 * g, whereafter the sediment was washed out with analar water. The radionuclide concentration of the liquid phase was measured on the Ge(Li) detector.

3) Fraction 2 : When the "readily exchangeable" fraction has been removed, it is assumed that the rest of the contamination can only be released by the application of agents which dissolve and bring about physico-chemical alterations of the form of the contamination or cause a destruction of the structure of soil constituents. A moderate reducing agent was therefore chosen for the second extraction. Here, $\text{NH}_2\text{OH}\cdot\text{HCl}$ in 25 vol % HAc has been shown to be a very efficient reagent for extractions of reducible metal oxide phases (Tessier et al. 1979). Therefore, the second extraction was performed with 20 ml of a 0.04 M solution of this reagent (at pH 2). The suspension was stirred rapidly for 6 hours at 80 C, before the separation and fractional radionuclide measurement were performed as described under fraction 1.

4) Fraction 3 : The third fractionation was performed with hydrogen peroxide which oxidizes the organic fraction of the soil, thereby releasing the radionuclides bound to this fraction. However, it has been pointed out that the oxidation by this method may not be entirely complete, and the addition of sodium pyrophosphate has actually been found to increase the released organic fraction in certain soils (Calcinaï & Sequi 1977). The procedure used in this fractionation was to add 15 ml of 30 % H_2O_2

(adjusted to pH 2 with HNO_3) to the residue from the second fractionation, and the suspension was again stirred rapidly for 6 hours at 80 C. Hereafter 5 ml of 3.2 M NH_4Ac in 20 vol. % HNO_3 was added in order to prevent resorption of released contaminants onto the oxidized sediment, and the suspension was stirred for another 30 minutes at room temperature, before the separation and spectral analysis, which were again performed as described above.

5) **Fraction 4** : A rather large fraction of the more persistently bound radionuclides has been proved extractable by treatment of the soil with more concentrated (7 M) HNO_3 . It is assumed that this effect is a result of a partial break-down of the clay structure. However, the results showed that a quite large fraction of the contamination was in fact not released by this method. Presumably, this pertains to the extremely strong collapsed interlayer fixation of cations in certain clays. The procedure for the release of this fraction was to add 20 ml 7 M HNO_3 to the residue from 3rd fractionation and stir for 6 hours at 80 C, before separation.

6) **Fraction 5** : This fraction is simply the strongly bound residue after the fourth fractionation. A calibration on the gamma spectrometer made it possible to compare the radionuclide content of this solid phase with that of the liquid phases.

It must be emphasized that the results of these speciation tests to some degree reflect reagent specific fractionations, although similar figures have been presented for humic soils for a speciation method with different extraction procedures (Livens & Baxter 1988), (Cook et al. 1984). These references also revealed that fallout Ru-106 in Cumbrian soils was only measureable in soils of great humic content, and that the organic chelate bound caesium fraction was as large as ca. 15 % in pasture soil.

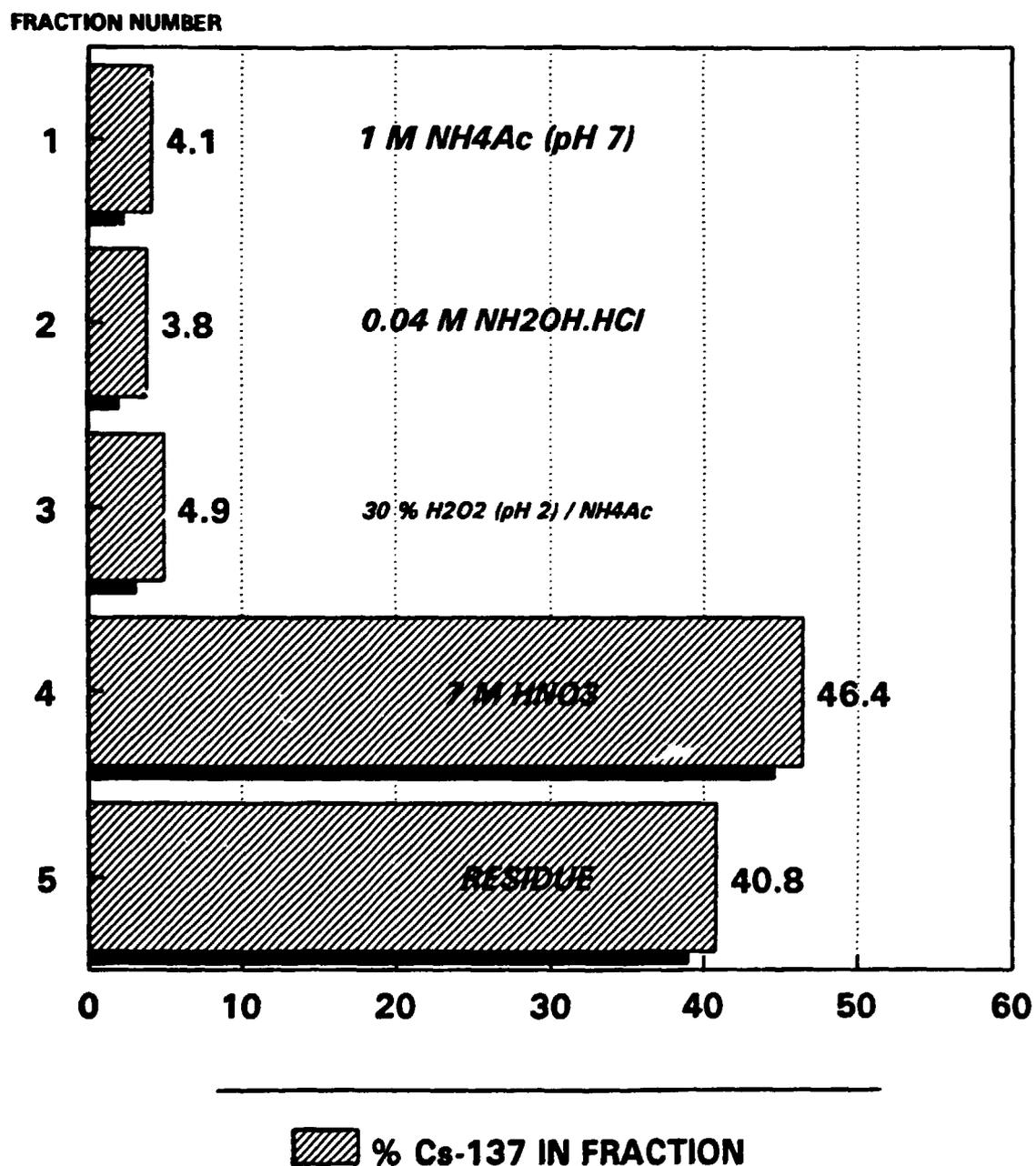
The results of the speciation experiments are shown in figures 3 - 7. In order to check the reproducibility of the method, the experiment presented in figure 3 was performed in triplicate. The results were identical within a 10 % margin. The correspondence between the results for Cs-137 and Cs-134 in the same profile (figures 3 and 7) can be seen as a check on the spectrometer accuracy.

One evident result of the tests is that the caesium is more weakly bound in the lower layers of the profile than in the top. Figures 3, 4, and 5 show the fixation in the top 0.8 cm layer, a layer 1.8 - 2.4 cm down in the profile, and a layer at the depth of 6.8 - 8.4 cm, respectively. As the depth increases, the exchangeable and easily dissolvable fractions increase. In the top layer, only ca. 13 % of the caesium was removed in the first 3 extractions, whereas in the 6.8 - 8.4 cm deep layer about 34 % was removed in these. This might be expected, as it is known that soil fixation of cations becomes more firm with time.

Figure 3.

SPECIATION ON CHERNOBYL SOIL

PERCENTAGE Cs-137 IN EXTRACTIONS No. 1-5

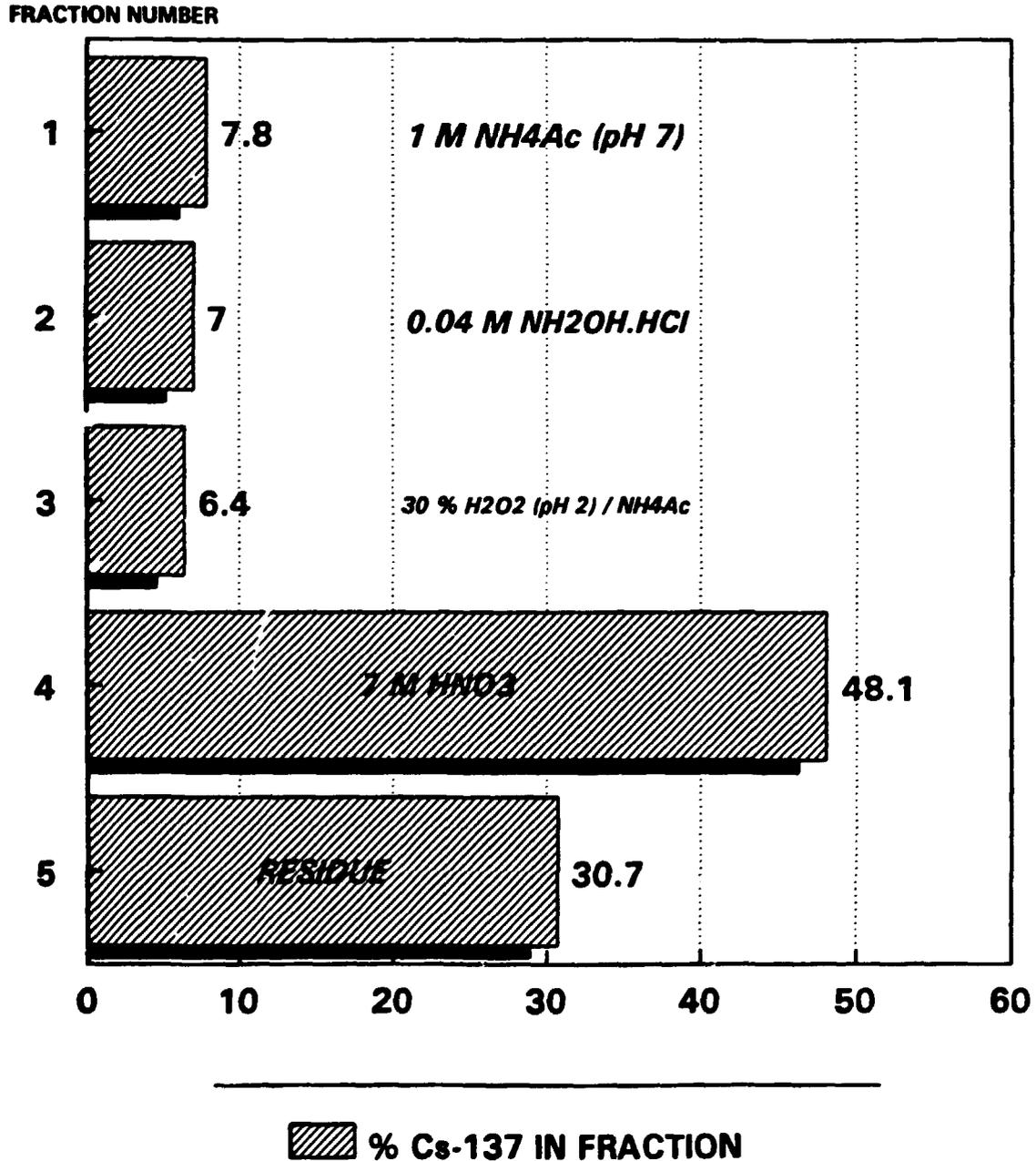


UPPER 0.8 CM LAYER CONTAINING 72.4 % OF THE Cs-137 IN THE PROFILE

Figure 4.

SPECIATION ON CHERNOBYL SOIL

PERCENTAGE Cs-137 IN EXTRACTIONS No. 1-5

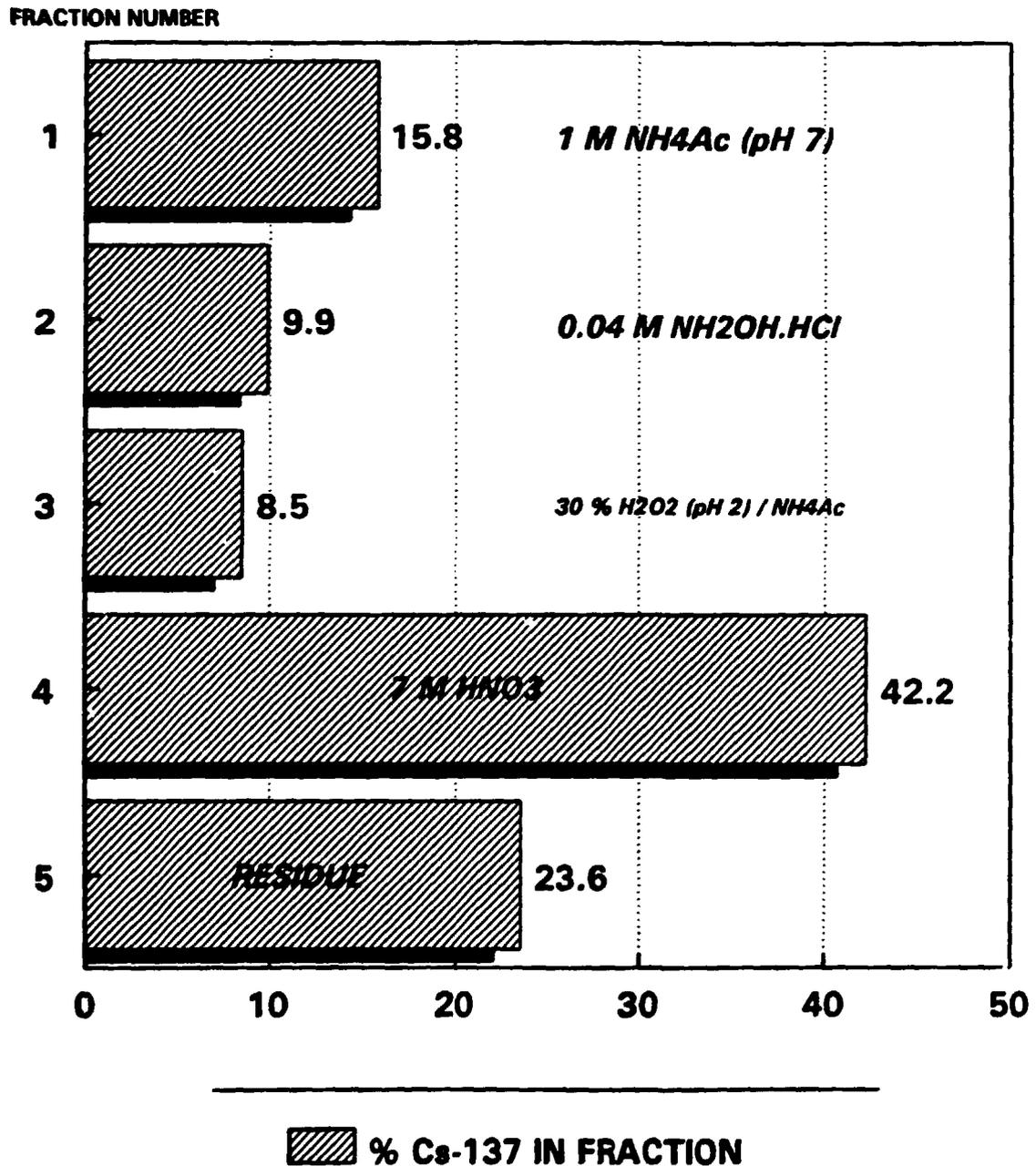


1.8-2.4 CM DEEP LAYER CONTAINING 3.0 % OF THE Cs-137 IN PROFILE

Figure 5.

SPECIATION ON CHERNOBYL SOIL

PERCENTAGE Cs-137 IN EXTRACTIONS No. 1-5

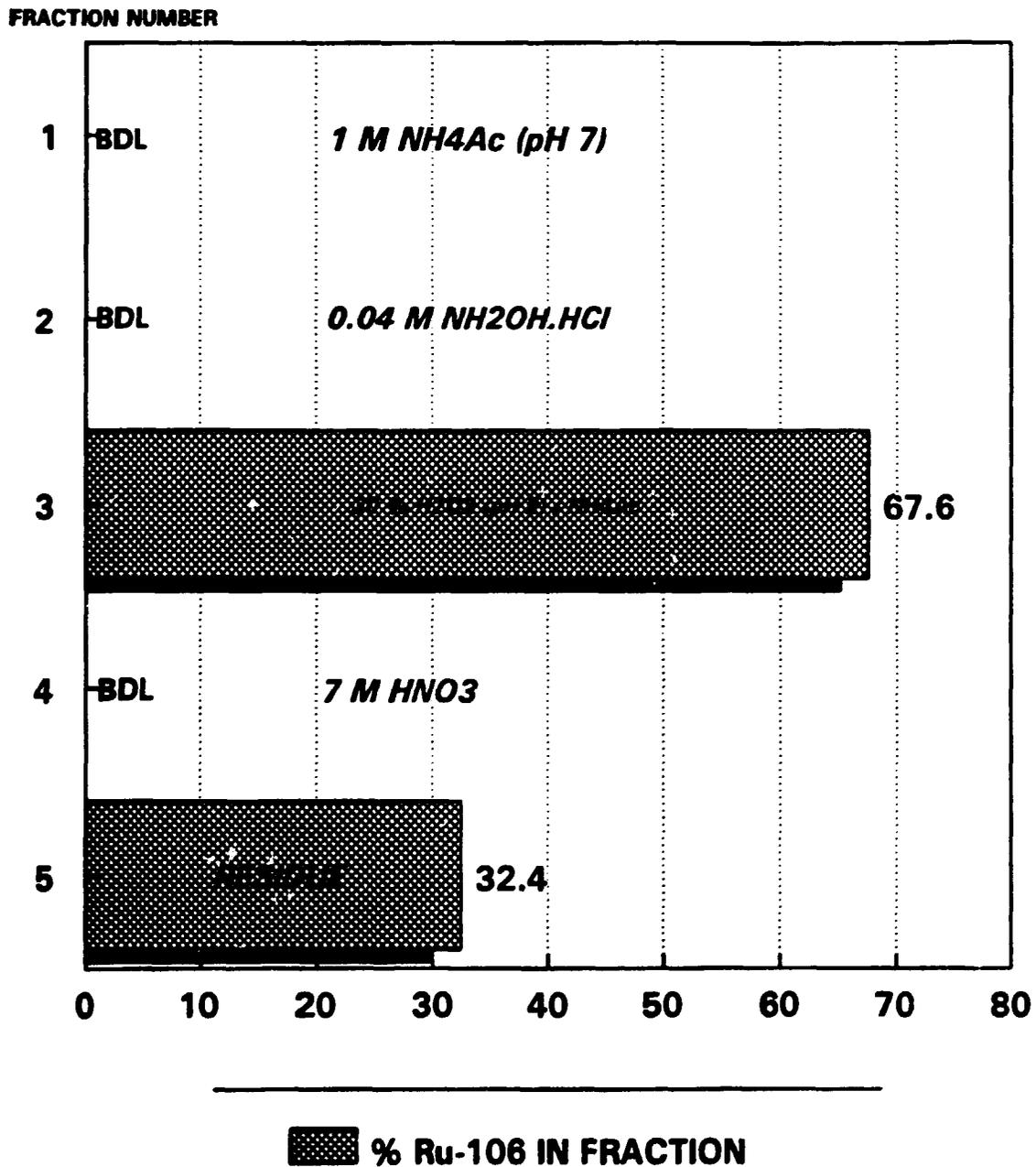


6.8-8.4 CM DEEP LAYER CONTAINING 0.4 % OF THE Cs-137 IN PROFILE

Figure 6.

SPECIATION ON CHERNOBYL SOIL

PERCENTAGE Ru-106 IN EXTRACTIONS No. 1-5

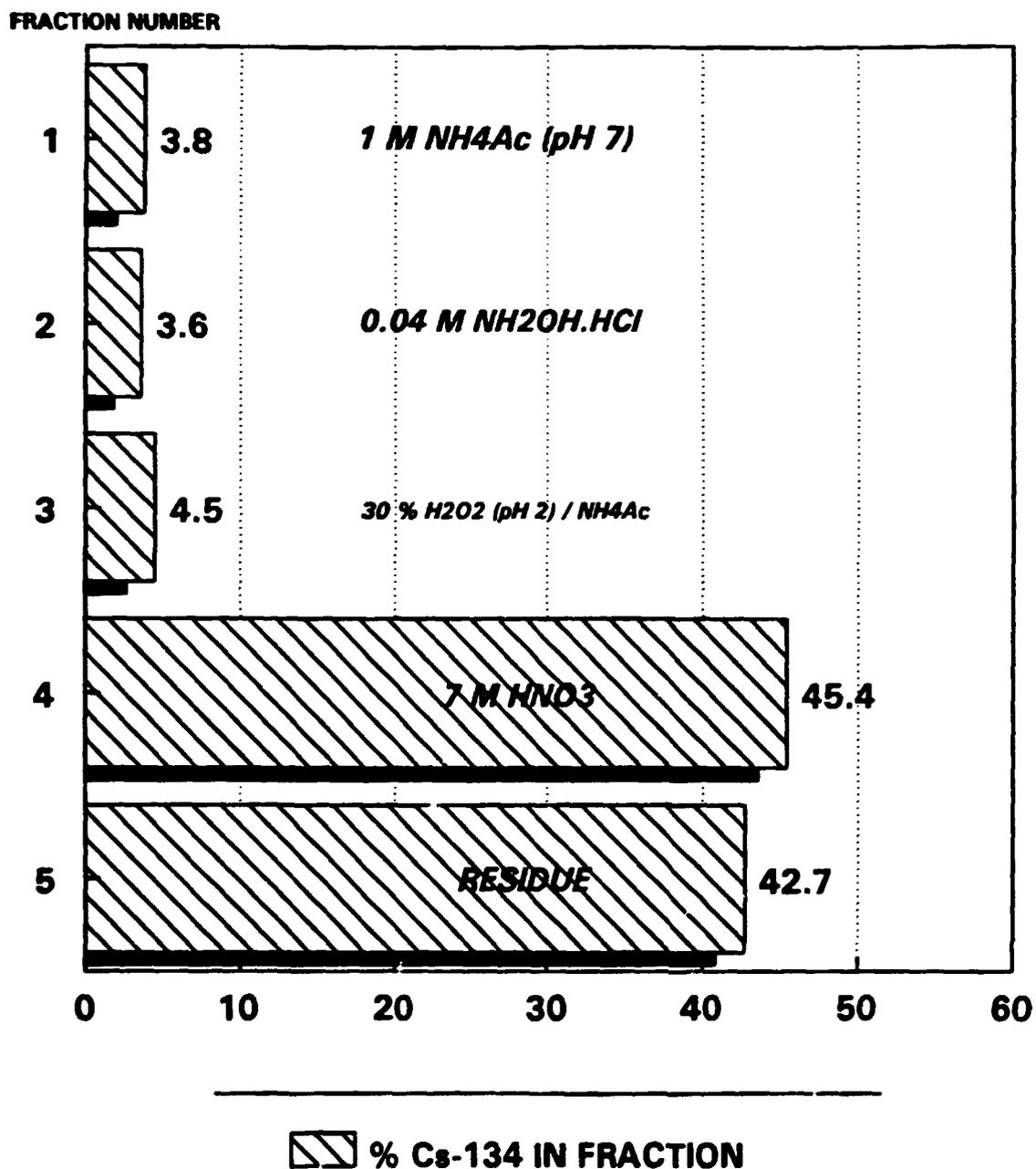


UPPER 0.8 CM LAYER CONTAINING 43.7 % OF THE Ru-106 IN THE PROFILE

Figure 7.

SPECIATION ON CHERNOBYL SOIL

PERCENTAGE Cs-134 IN EXTRACTIONS No. 1-5



UPPER 0.8 CM LAYER CONTAINING 71.2 % OF THE Cs-134 IN THE PROFILE

For clay minerals, the "irreversible" collapsed c-spacing fixation may take years to complete (Squire & Middleton 1966). This may be reflected in the smaller fraction of the extremely firmly fixed residue in the deeper soil layers.

The speciation analysis for ruthenium gave a very different result (fig. 6). Ruthenium levels could only be detected in the upper layers of the soil, where the humic content is very high. Only the fraction bound to organic material and the very strongly bound residual fraction were measureable. About two thirds of the measured ruthenium was found to be associated with the organic fraction. It must be stressed that the numbers given are percentages of the measureable ruthenium. The term "Below Detection Limit" should be read as "less than 3 % of total Ru-106 in fraction".

As a general conclusion for the experiments it can be said that the fixation of the investigated radionuclides in the upper layers of the examined soils is so strong that non-aggressive reclamation procedures are unlikely to have any effect. However, only a very thin top layer needs to be removed to decrease the external dose from the contamination substantially.

3. The Efficiency of Reclamation Procedures for Gardens.

As already mentioned in the introduction the currently recommended method for the reduction of the external exposure from contaminated garden soil is to dig it to the depth of one spit. This should reduce the external dose to ca. 17 % of its initial value.

As an alternative to this method the application of several different types of "enclosing", strippable coatings has been investigated.

The following coatings were tested in small scale experiments :

3.1. PVA coating.

This is a modified version of a method for decontamination of smooth surfaces (Robev et al. 1966). The agent, as it was used, was based on a 10 % polyvinyl alcohol solution in water to which 20 % ethyl alcohol, 1 % EDTA (for chelate formation) and 0.02 % sodium carbonate was added.

The contaminant (Cs-137) was first sprayed onto the surface in a fine mist of water (corresponding to < 0.1 mm rain). The thick PVA liquid was then applied by brush onto a freshly contaminated bare soil surface of about 0.03 m², although the recommendation in large

scale would be to spray it on. When the coating had been allowed to dry for a couple of hours it was easily peeled off, taking with it the upper few millimetres of soil. On grassed areas it would be recommended first to cut the grass as short as possible, to enhance penetration of the fixative.

As the activity at this point is embedded in the film, the resuspension and "smearing" hazard will have been reduced greatly.

The total amount of waste generated from an area of 0.03 m² was approximately 100 g.

The gamma radiation from the contaminated surface was measured by a lead shielded 15.6 % efficiency Ge(Li) detector system before and after the application of the agent. It was revealed that 93.8 % of the activity had been removed with the coating. Of course the effect depends on the penetration of the contamination into the soil, but in most cases the major part of the activity will be found in the uppermost millimetres directly after deposition. The penetration of the PVA liquid can be adjusted by alterations to the viscosity. Likewise, the length of the polymers can be changed by the addition of glycerins or starches.

PVA is to some degree dissolvable in water and biodegradable over longer periods of time (Skriver 1990).

It is important to estimate the cost of performing the operation. The most expensive of the chemicals is the PVA powder. In large amount this is sold for about 2.50 ECU per kg, and it is available in very large amounts. This means that the price per square meter is about 0.20 ECU. The total cost of chemicals should therefore be less than 0.40 ECU/m². In addition there is the cost of transport. From a garden area of 200 m², the amount of generated waste is $200 \text{ m}^2 * 0.1 \text{ kg} / 0.03 \text{ m}^2 = 0.7 \text{ tons}$. If this waste is transported on large trucks, the costs per reclaimed square meter will be relatively small.

Then there is the matter of the payment to the people performing the operation. It should not be necessary to remove the surface layer by hand. The coated surface could be "lifted" off with a shovel, thereby reducing the exposure to the worker. But the procedure would be less demanding than it would be to dig the soil to the depth of one spit, which at present is the recommendation. The cost for that countermeasure was estimated to ca. 1 ECU per square meter (Gjørup et al. 1982). So on an economical scale the PVA procedure seems practicable.

It must however be stressed that under extremely wet conditions it may not be possible for the film to dry sufficiently.

Similar coatings have previously been used as radiation fixatives

to facilitate the clean-up in the more sandy parts of the Chernobyl area (Komarov 1989).

3.2. Lignosol BD coating.

Lignosol BD, as one of the commercially available forms of lignin is called, is one of many byproducts of technical processes such as wood pulping or wood hydrolysis, for instance in paper mass production. The amount of lignin "waste" from such processes amounts to ca. 40 million tons per year (Schweers & Vorher 1977).

In biomass lignin is generally second in quantity after cellulose (Kononova 1951). The lignin molecule is a large and complex polymer, which is very efficient in forming chelates with cations, even with monovalent ions like caesium. The humic cation fixation in soils is believed to be controlled by fragments of lignin-like molecules, which, probably due to chelate formations, decompose at a much slower rate than for instance cellulose. Therefore, quite large concentrations of these molecules can be accumulated (Sørensen 1990).

However, by wood pulping processes, the lignin molecule undergoes some condensing reactions and is usually transformed into sulphuric derivatives. Thereby, the lignin molecule will have been made somewhat less reactive, although its binding abilities are still sufficient to be utilized for numerous purposes.

Among the applications featured by the English distributors (Barron 1990) are :

Binding of pelletized metal ores, clay and ceramics products. Dust control / coating of coal, adhesives in linoleum, cement and paper cores, stabilization of wax emulsions and molasses, and extension of resins. Control of dust on dirt roads.

In this connection the latter is probably the most interesting. The lignin byproducts are in wide use for this purpose in the States, and for instance also in Sweden, where the lignin waste is a big problem. The use of calcium lignosulphonate as a radiation fixative has been strongly recommended (Tawil & Bold 1983), as it is non-toxic and biodegradable, and can be washed off with water before it dries and afterwards with an oil solvent and light scrubbing. It is diluted with water and applied with a water spray truck. Furthermore, it is of course inexpensive. In the United States it is sold under the name of "lignosite", and the cost is given to be 20 cents per gallon in 1983 prices.

It is available as a dry product in 25 kg bags, and in liquid form in 10-20,000 gallon tank cars (Barron 1990).

Lignosol BD was tested in a thick water solution, in the same way as described above for the PVA coating. The polymerization, and thereby the strength of the removed surface layer, was not quite as good as that of the PVA coating, but the top few millimetres were still easily removed. The surface should be removed before the agent dries completely, as it will be less brittle. The mass of the removed layer was approximately the same as that of the PVA layer.

Like the PVA coating, the lignin coating should not be used under extremely wet conditions.

With the lignosol coating in the small scale test, 93.9 % of the caesium was removed with the coating on bare soil, while 71.5 % was removed from a cut grassed surface.

The conclusions on comparison with the PVA coating must be that the lignin coatings will have less strength, but also be much cheaper.

3.3. Other coatings

"Liquid envelope" - a peelable protective plastic coating manufactured by Cufadan A/S in Copenhagen for special surface treatments was also tested in the same way as the PVA coating. The agent was applied by brush, although it could have been done by spraying. Here, 94.7 % of the caesium contamination was removed. The coating had good strength, although the PVA coating seemed more suited for the purpose. Also, the "liquid envelope" coating would be much more expensive.

A number of other types of coatings were examined with less success. Resins possess good adhesive characteristics, and are relatively cheap (ca. 1 ECU / kg) and available in large amounts. They are available in many forms, tailored for different purposes. However, it was not possible to make a sufficiently strong film out of any of the two investigated resin types Staybelite Resin E and Staybelite Ester 3E from Hercules.

It was also impossible to gain sufficient strength in films of "krystal fixativ", a commercially available spray mixture of ethyl alcohol, PVP, and resins. A Valvoline "Tectyl" lacquer spray was also investigated without sufficient success.

4. Summary and Conclusions

The fixation effect of various soil constituents on the radioactive ions Cs-134, Cs-137, and Ru-106 in the Chernobyl debris was investigated.

The investigations were carried out on soils from heavily contaminated areas in the Soviet Union and in Sweden.

The results of soil profile samplings indicated a much stronger fixation of caesium than of ruthenium ions.

Speciation tests showed that most of the contamination (specially the caesium) in the upper layers was remarkably firmly fixed. Much of the contamination was not even leached by concentrated nitric acid. Another result of the speciation experiments was that in contrast to the caesium, most of the ruthenium was bound in weaker organic complexes.

The speciation tests also showed a weaker fixation of caesium in the lower soil layers, which presumably is due to the time factor of the irreversible caesium fixation in clays.

Some new methods for decontamination of urban soil were suggested. The methods were based on the application of strippable coatings of different types.

Small scale experiments gave good results for a contamination which was performed with very little water.

The most efficient of the examined coatings were based on PVA and calcium lignosulphonate, respectively.

The coatings could also be used in clean-up procedures for non-radioactive pollutants.

5. Acknowledgments

The author wishes to thank the following persons for their co-operation and good advice :

Miriam A. Byrne, Nuclear Power Section, Mechanical Engineering Department, Imperial College, London SW7 2AZ, UK.

Henrik Hougård and Henrik Prip, Ecology Section, Risø.

6. References

- Allison F.E., Soil Organic Matter and its Role in Crop Production, U.S. Department of Agriculture, D.C., USA, Elsevier, 1973.
- Barron, D., Roy Wilson Dickson Ltd., Chester, England, personal communication, 1990.
- Brady, N.S., The Nature and Properties of Soils, 8th edition, Macmillan Publishing Co. inc., New York, 1970.
- Bunzl, K., Schimmack, W., Kreuzer, K., Schierl, R., The Migration of Fallout ^{134}Cs , ^{137}Cs and ^{106}Ru from Chernobyl and of ^{137}Cs from weapons testing in a forest soil, Z. Pflanzenernähr. Bodenk., 152, 39-44, 1989.
- Calcinai, M. and Sequi, P., Contribution of Organic Matter to Cation-Exchange Capacity of Soils, in proceedings of the symposium on soil organic matter studies, Braunschweig, 6-10 Sept., 1976, IAEA proceedings series, Vienna, 1977.
- Chaberek, S. and Martell, A.E., Organic Sequestering Agents, A discussion of the chemical behaviour and applications of metal chelate compounds in aqueous systems, Wiley, New York, 1980.
- Cook, G.T., Baxter, M.S., Duncan, H.J., Toole, J., Malcolmson, R., Geochemical Association of Plutonium in the Caithness Environment, Nuclear Instruments and Methods in Physics Research, 223, 517-522, 1984.
- Duff, R.B., Webley, D.M., Scott, R.O., Solubilization of Minerals and Related Materials by 2-Ketogluconic Acid-Producing Bacteria, Soil Science, 95, 105-114, 1963.
- Gjørup, H.L., Jensen, N.O., Hedemann Jensen, P., Kristensen, L., Nielsen, O.J., Petersen, E.L., Petersen, T., Roed, J., Thykier-Nielsen, S., Heikel Vinther, F., Warming, L., Aarkrog, A., Radioactive Contamination of Danish Territory after Core-melt Accidents at the Barsebäck Power Plants, Risø National Laboratory, Risø-R-462, 1982.
- IAEA Report on Methodology and Technology for Cleanup of Very Large Areas After a Nuclear Accident, Editor : M.A. Feraday, IAEA 7857w, 1987.
- Komarov, V.I., Radioactive Contamination and Decontamination in the 30 km Zone Surrounding the Chernobyl Nuclear Power Plant, in proceedings of the IAEA symposium on Environmental Contamination following a major nuclear accident, Vienna, 16-20 Oct., 1989.
- Kononova, M.M., Soil Organic Matter - Its Nature, Its role in Soil Formation and Soil Fertility, Pergamon Press, Oxford, 1961.

Livens, F.R. and Baxter, M.S., Chemical Associations of Artificial Radionuclides in Cumbrian Soils, J. Environ. Radioactivity, 7, 75-86, 1988.

Nishita, H. and Essington, E.H., Effect of Chelating Agents on the Movement of Fission Products in Soils, Soil Science, 103, 168-176, 1967.

Robev, S.K., Sumerska, T.B., Todorov, S.L., Kristanov, D.K., Marinova, I., A Method of Decontaminating Radioactively Contaminated Surfaces by Film, in proceedings of the Symposium on Practices in the Treatment of Low and Intermediate Level Radioactive Waste, IAEA, Vienna, 1966.

Roed, J., Andersson, K.G., Sandalls, J., Reclamation of Nuclear Contaminated Urban Areas, Presented at the BIOMOVs Symposium and Workshop on the Validity of Environmental Transfer Models, Stockholm, 8-12 Oct., 1990.

Route, G., Biotechnology, B353, Harwell Laboratory, Oxon, England, personal communication, 1990.

Salbu, B., Bjørnstad, H.E., Krekling, T., Lien, H., Riise, G., Østby, G., Determination of Physicochemical Forms of Radionuclides Deposited after the Chernobyl Accident, in proceedings of the IAEA Symposium on Environmental Contamination following a Major Nuclear Accident, Vienna, 16-20 Oct., 1989.

Schnitzer, M. and Khan, S.U., Soil Organic Matter, Elsevier, 1978.

Schweers, W.H.M., and Vorher, W., Possibilities of an Economic and Non-Polluting Utilization of Lignin, in proceedings of the IAEA Symposium on Soil Organic Matter Studies, Braunschweig, 6-10 Sept. 1976, IAEA Proceedings Series, 1977.

Skriver, K., Hoechst Chemicals Danmark A/S, personal communication, 1990.

Squire, H.M. and Middleton, L.J., Behaviour of Cs-137 in Soils and Pastures - a Long Time Experiment, Radiation Botany, 6, 413-423, 1966.

Sørensen, A., Risø Agricultural Dept., personal comm., 1990.

Tawil, J.J. and Bold, F.C., A Guide to Radiation Fixatives, Pacific Northwest Laboratory, Richland, Washington 99352, USA, PNL-4903, 1983.

Tessier, A., Campbell, P.G.C., Bisson, M., Sequential Extraction Procedure for the Speciation of Particulate Trace Metals, Analytical Chemistry, Vol. 51, No. 7, 844-851, 1979.

<p>Title and author(s)</p> <p>THE CHARACTERIZATION AND REMOVAL OF CHERNOBYL DEBRIS IN GARDEN SOILS.</p> <p>Kasper G. Andersson</p>	<p>Date January 1991</p> <p>Department or group Ecology Section</p> <p>Groups own registration number(s)</p> <p>Project/contract no. 02369-00 - ECO</p>
<p>Pages 26 Tables 1 Illustrations 7 References 25</p>	<p>ISBN 87-550-1699-5</p>
<p>Abstract (Max. 2000 char.)</p> <p>Severe nuclear accidents such as the one in Chernobyl in 1986 may give unacceptably high external radiation levels, which even in the late phase may make a resettlement of an evacuated population impossible unless action is taken to decrease the exposure. As the urban land areas to be reclaimed may be very large the cost of the dose reducing countermeasure to be used may be an important factor. In the Chernobyl debris the most important radionuclides concerning the long term external radiation were found to be Cs-137, Cs-134, and Ru-106. Therefore, the aim of this work is to investigate the behaviour of these radionuclides in garden soils, and on this background to examine cost-effective methods by which a reduction of the dose from such areas to people living in urban or sub-urban environments can be achieved.</p> <p>The fixation of the radioactive cations in soil was investigated by means of soil profile sampling, soil texture analysis, and speciation experiments. It was found that most of the Chernobyl fallout caesium was extremely firmly fixed. Much of the ruthenium was more loosely bound, to organic material.</p> <p>The cost-effectiveness of some dose reducing countermeasures was examined on the background of small scale tests. Here it was found that about 95 % of the activity could be removed with peelable fixatives based on PVA or lignin.</p>	
<p>Descriptors - INIS</p> <p>CATIONS; CESIUM 134; CESIUM 137; CHEMICAL STATE; CHERNOBYLSK-4 REACTOR; COST BENEFIT ANALYSIS; DECONTAMINATION; FALLOUT DEPOSITS; ION EXCHANGE; LAND RECLAMATION; LIGNIN; MELTDOWN; PVA; RUTHENIUM 106; SOILS; URBAN AREAS</p> <p>Available on request from Riso Library, Riso National Laboratory, (Riso Bibliotek, Forskningscenter Riso), P.O. Box 48, DK-4000 Roskilde, Denmark. Telephone +45 42 37 12 12, ext. 2268/2269. Telex: 43116, Telefax: +45 46 75 66 27</p>	

Available on exchange from:
Risø Library
Risø National Laboratory,
P.O. Box 49, DK-4000 Roskilde, Denmark
Phone + 45 42 37 12 12, ext. 2268/2269
Telex 43116, Telefax + 45 46 75 56 27

ISBN 87-550-1699-5
ISSN 0418-6435