WASTE MANAGEMENT OF CONTAMINATED CROPS AFTER A LWR ACCIDENT

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ABSTRACT

Within the program on rehabilitation of rural surfaces and soils after a nuclear accident (RESSAC), investigations were undertaken to study the possibilities of effective waste management of dry and fresh radioactively contaminated crop biomass considering mainly biodegradation processes. The basic idea of the study was to find the simplest and most effective methods and/or means for a fast and low cost treatment of contaminated biomass under field conditions. The present study gives the first results on effective decontamination as well as on reduction of biomass (volume and weight). For the resulting radioactive liquid end products, easy treatments with approved methods and means are proposed in order to facilitate safe disposal.

INTRODUCTION

After a nuclear accident, clearing is a common decontamination method for agricultural fields, vacant lands, and wooded areas (1). Work performed within the European RESSAC program (Rehabilitation of Soils and Surfaces after an Accident) for agricultural fields proved that plants like ripening wheat can intercept about 80% of the total deposited dry particles of 1 μm in diameter (2). Clearing wet and dry herbaceous plants with a silage harvester would be an efficient method if such contaminated biomass can be managed in order to avoid creation of new source terms. Considering, for example, wheat has a dry biomass production of 2 kg/m^2, the waste management for thousand hectares becomes a complex problem for which feasible solutions should be foreseen.

Caesium and strontium are the main radionuclides present in the environment after an accident. Since accidents will necessitate emergency countermeasures, technical means for management must be as simple as possible, excluding, however, such options as chemical treatment, incineration, not adapted to huge quantities of contaminated biomass. Natural biodegradation of the piled biomass will occur in any case without human intervention which could lead to partial decontamination of the biomass if efficient processes can be successfully managed. Some laboratory experiments have been carried out in order to get an idea of the expected results and the conditions to practice in a decontamination schedule, before the final validation trials under field conditions.

EXPERIMENTAL

Two sets of experiments were performed with cress, haricot, and wheat in order to study microbial decomposition and decontamination of fresh and dry organic matter. Different natural conditions of growth stage and water content of the plants were simulated aiming at the most appropriate and efficient methods for the treatment of radioactive contaminated organic wastes.

The plant species were grown on a hydroponic system spiked with Cs-134 and Sr-85 resulting in a faster and higher contamination by root-uptake than in soil cultures. This option was also chosen because contamination of crops by root-absorption represents the strongest possible immobilization of radionuclides in plants and normally results are then the most difficult to be released.

Small Scale Pilot-Experiment

Aliquots of fresh cress biomass (100 g) and haricot (20 g) were harvested after one month of growth and accumulation of radioactivity. A part of the biomass was oven dried at 333K up to constant weight. Both dry and fresh samples were subdivided into three groups (each initially weighing 100 g when fresh) and subjected to aerobic and anaerobic biodegration in polythene containers (500 cm^3). No inoculation of the biomass with special microorganism strains was applied to speed microbial processes.

Since accidental radioactive contamination of crops by fall-out may happen in all seasons and, hence, at all growth stages of the plants affected, trials were undertaken to simulate various humidity conditions of the biomass by either using fresh material or by adding different aliquots of water to the dry biomass, i.e., multiples of the initial fresh weight. Parallel sets of dry biomass samples received 50, 100 and 200 cm^3 of tapwater in both aerobic and anaerobic conditions while for the fresh biomass only to one set of samples was tapwater added, the rest remained without any amendment.

Bigger Scale Experiment

This work was aimed at defining the best conditions to be used in the fields after an accident for the management of piles of contaminated biomass weighing hundreds of tons. Because such big quantities cannot be handled in a laboratory and experiments with several grams are hardly representative of reality, kg scale trials have been made as close as possible to the field conditions. Aliquots of fresh biomass of cress (10 kg) and wheat (3 kg) were harvested at ripeness.

About half of the cress sample and all of the wheat were oven dried at 333K. The dry samples of cress and wheat received 3,000 cm^3 and 6,000 cm^3 of tapwater, respectively, and were subjected to anaerobic biodegration. No inoculation of the biomass with microorganisms was applied.

Measurements

Measurements of produced and recovered liquids and released radioactivity by simple percolation were carried out at regular time intervals in order to evaluate the degree of biodegradation and decontamination of the biomass.

The gamma-emission spectra of Cs-134 and Sr-85 were measured with a Ge-detector connected to a multichannel analyzer (TN 7200).

At the end of the experimental period, the reduction of the biomass in terms of weight and volume was calculated as a percentage of the initial measurements.

Leaching of Radioactivity

Trials to improve the reduction of the remaining radioactivity in the biomass after simple percolation were carried out using different agents such as the addition of water, NaOH, NH4-acetate, Ca-sulfate, K-chloride, Fe-chloride, and EDTA at different concentrations from 10 to 40 mg/cm^3. After the drainage of the initial water, the salt solutions were used to reimmerse the solid biomass at the ratio of 1 cm^3 of solution per gram of biomass. Parallel to chemical leaching, mechanical carrying of the radioactivity by small particles was simulated by sequential water washing of the solid biomass. The management of released liquids at the end of the process takes on an important part in the whole method's success: Therefore, the soluble and the dissociated radionuclide fractions in released liquids were determined by centrifugation at 11,000 g, filtration through membranes of 0.01 μm and contact with strong cation-exchange-resins in a batch system.

RESULTS AND DISCUSSION

Aerobic Versus Anaerobic

Within the various conditions tested considering aerobic and anaerobic conditions, dry and fresh biomass and two different crop species (cress, haricot), microbial biodegradation proved best under anaerobic conditions.

The release kinetics of caesium and strontium expressed as the percentage of the initial activity under aerobic and anaerobic conditions starting with dry plant material (cress) are shown in Fig. 1.

For both caesium and strontium, anaerobic digestion resulted in the highest release percentages, i.e., about 50% was transferred to the liquid phase, produced during the process and recovered by percolation only. The efficiency of the aerobic process of degradation was about 20% lower, reaching maximum values of about 35% as well for caesium as for strontium.

The different release behavior of Cs and Sr either in aerobic or anaerobic conditions could be explained by leaching experiments with water. Leaching of caesium and strontium from dry biomass occurs fast within the first two hours of contact of water with the dry biomass. In anaerobic conditions, about 40% of the caesium and 17% of the initially present strontium is leached during the next two hours reaching maximum values for caesium and strontium of 45% and 20%, respectively. This is reflected in the initial values of Fig. 1, where starting points of release of caesium and strontium coincide with those values.

After the initial leaching, the release behavior of caesium and strontium were found to be quite different. The overall release for caesium increased only slightly with time reaching a final maximum value of 50% after 43 days while the initially low release of strontium (17%) was strongly affected by the degradation process and increased rapidly with time to a final value of about 50%: Thus, similar release maxima were found for caesium and strontium, yet 15-20% lower for aerobic conditions than for anaerobic digestion. The best efficiency of anaerobic conditions may be due to the accumulation of carboxylic anions produced by anaerobic microflora able to form soluble salts with cations like caesium and strontium. In aerobic conditions, these anions are more quickly decomposed into carbon dioxide and then lacking radionuclide solubilization. This is well described in the literature (3).

Effects of Humidity and Added Water Volume on Biodecontamination

Dry biomass of most of the corn crops (wheat, barley, rye and maize, etc.) will hardly start quick decomposition processes when piled up because of the lack of humidity essential for good growth of microorganisms. Therefore, additions of water to the dry biomass were considered to re-establish different water contents in the biomass according to seasons and growth stages of the crops at the time of their premature harvest after radioactive contamination by fallout. Furthermore, natural water content in the biomass and additional water volume should enhance the microbiological degradation processes as well as favor the leaching of radioactivity from the biological structures. The experimental results confirmed that increasing water volume leads to an increase in the released radioactivity (Fig. 2). Water action seems a vector of the radionuclides which may be due to the equilibrium ratio between the solid and liquid phase. Up to the equilibrium condition limits, the concentration of the liquid phase remains constant and, therefore, the total activity of the liquid increases with the volume of liquid. For both radionuclides (Cs and Sr), pronounced effects of water

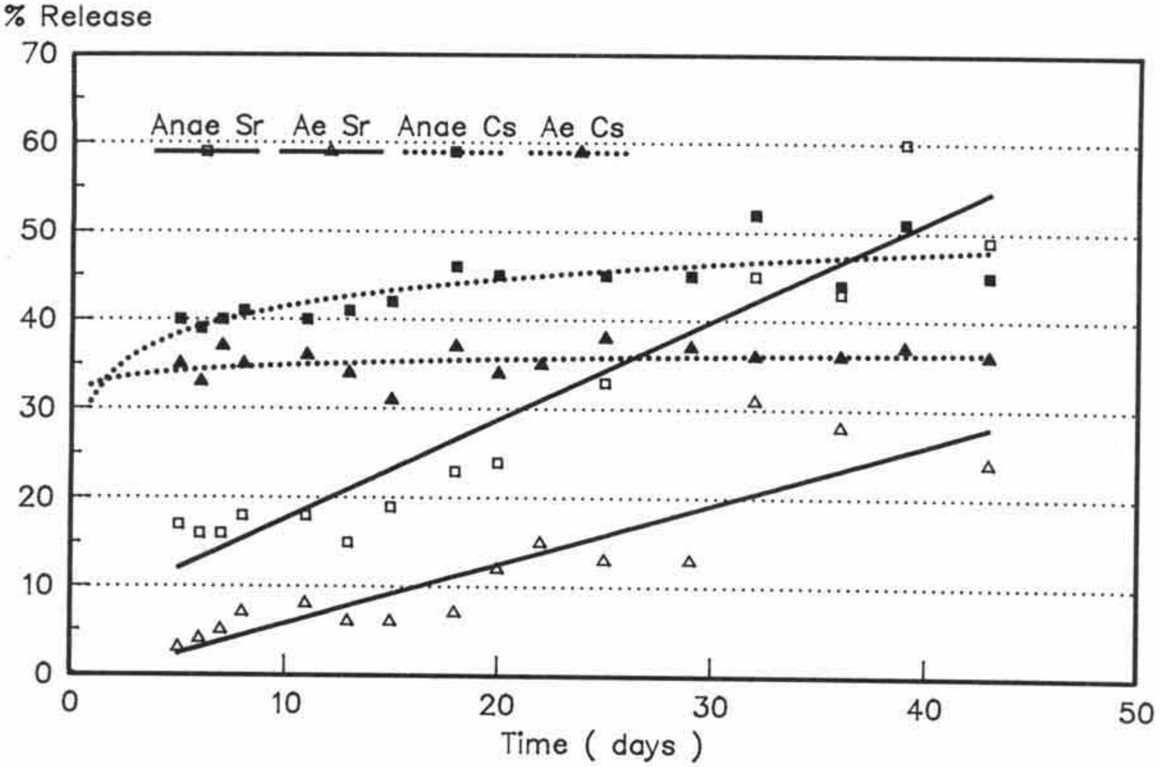

Fig. 1. Aerobic and anaerobic digestion of cress biomass and radionuclide release.

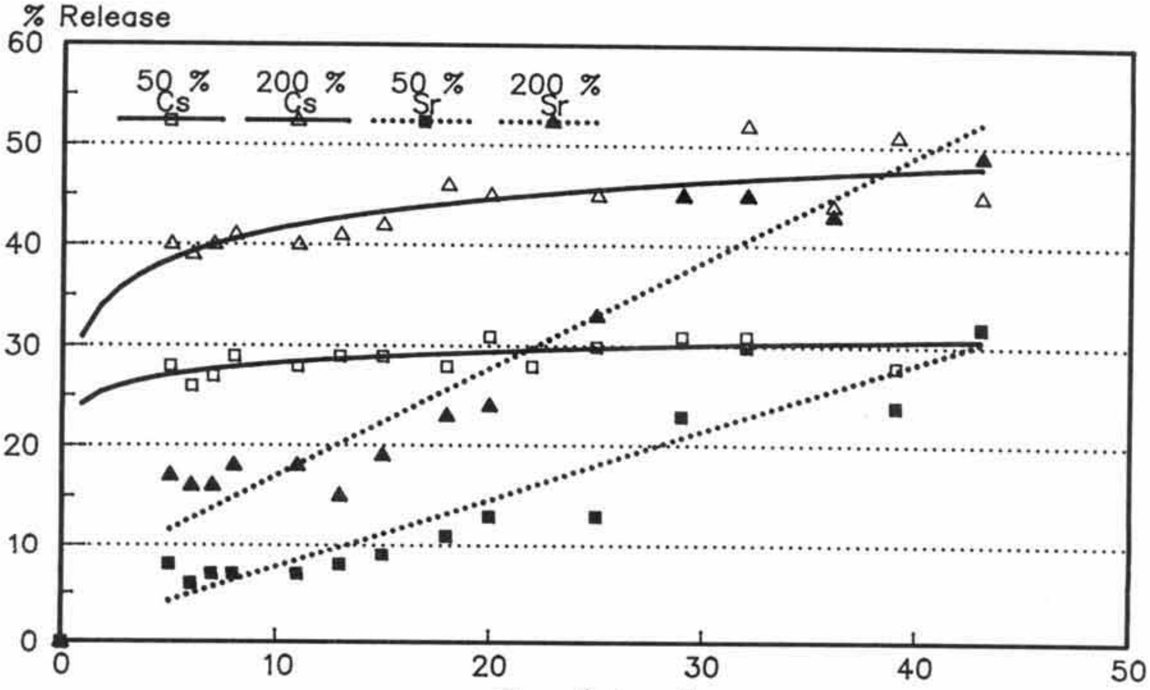

Fig. 2. Effect of water volume on decontamination effciencies in anaerobic digestion of contaminated dry cress.

addition on the overall release of radioactivity could be observed showing higher efficiencies for strontium than for caesium yet it resulted in the same decontamination levels in both conditions for Cs and Sr. Water addition may be a favorable condition for decontamination but the scale of the experiment must be taken into account. A minimum volume of water will have to be used in the fields in order to avoid the management of water weight and volume bigger than the biomass itself.

Effect of Experiment Scale and Plant Species

The percentage of total radioactivity released in the liquid (Kg-scale-experiment) was close to 50% for caesium, but did not exceed 25% for strontium (Fig. 3) instead of 50% in the gram-scale-experiment. These results show that it may be more difficult to recover the radioactivity due to strontium in the liquid phase when the amount of biomass increases. This may be due to the great affinity of strontium for organic matter already mentioned in the literature (4). This is confirmed by the results where about 95% of strontium and only 15% of caesium radioactivity in the released liquid was found in sediments after centrifugation at 11,500 g.

The small organic particles are sticking to the bigger particles in the pile and are not removed by a single drainage of the pile. This fact was not true in the gram-scale-experiment because the biomass had a higher surface to volume ratio.

With respect to the different plant species considered and their different biological structure, i. e., lignin-cellulose, the decontamination efficiency by anaerobic digestion was similar for caesium and differed for strontium showing lower values for fresh biomass than for the dry one (Fig. 3). This observation may be explained by the fact that in the fresh biomass, vital structures and compounds might have been intact after digestion and, therefore, could interact with strontium in binding and blocking it excluding decontamination.

Leaching of Radioactivity

Besides the initial leaching with pure water which resulted in 55% and 15% elution of Cs and Sr, respectively, within two hours, trials were performed to increase liquid release of radioactivity by replacement of water with saline solutions. The respective salt efficiency is given in Table I, which clearly shows that no compound gives significant results better than water alone.

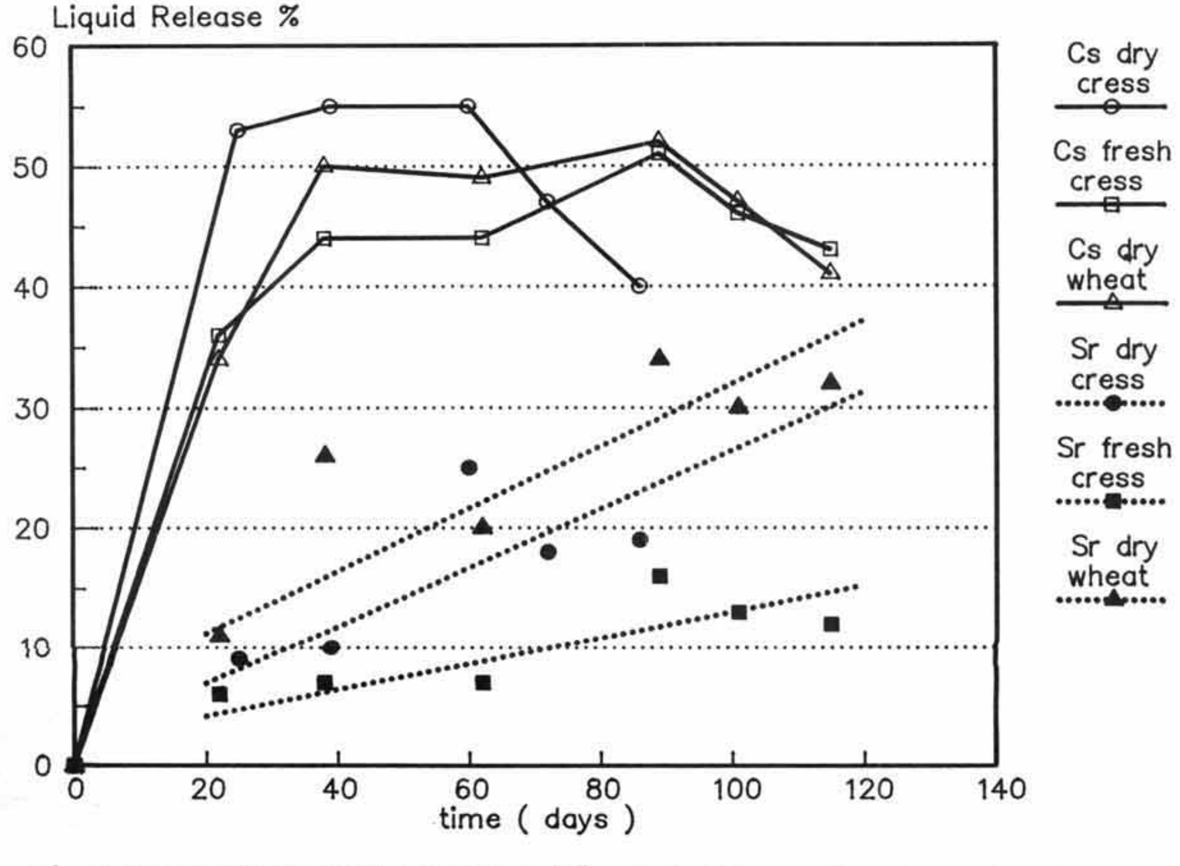

Fig. 3. Kinetics of radionuclide release from different plant biomass: Kg scale experiment.

TABLE I

Leaching Efficiency of Radioactivity from Contaminated Biomass

Solution and % Concentr.		Leached %	
		Cs	Sr
NH4SO4	1%	41	10
CaNO3	1%	44	12
NH4CO2	2%	51	28
FeC13	2%	52	22
NH4CO2	4%	52	21
Kc1	4%	53	20
NH4SO4	2%	53	12
dist.	H2O	55	15
NaOH	2%	60	14
EDTA	2%	60	27

Sequential water washing trials allowed to increase the simple immersion efficiency from 55 to 65% for caesium and from 15 to 57% for strontium. In case of sequential leaching, the release gain may be due to mechanical carrying by the water stream reaching a higher level for Sr than for Cs. Thus, an average leaching efficiency of radioactivity amounted to about 60% for the two nuclides. However, it is difficult, based on laboratory trials only, to evaluate the volume of water necessary for washing and leaching of radionuclides in a real size pile. Recycling of the leaching water stream may allow optimization of liquid waste production.

Balance and Behavior of Remaining Biomatter

The residual solid biomass and volume related to the dry or fresh initial state is given in Table II. The remaining liquid volume is related to the initial natural content (fresh cress) or supplied water (dry plants). According to these results, the mass and volume reduction seems to be dependant on both the plant type and the state of desiccation of the plant. Wheat has the most difficult plant matter to reduce. Since the anaerobic digestion leads to an important reduction in mass and volume, further field handling will concern smaller quantities than the initial wastes. Therefore, a better decrease of the contamination level would be more realistically attempted. The remaining radioactivity may be linked to cellulosic structures of the biomass. According to other studies in the RESSAC program, such an organic structure as carboxymethyl-cellulose can link 60% of caesium and 85% of strontium in an aqueous solution. Cellulose may occur and act as an immobilization structure of radionuclides. Potential enzyme decomposition of cellulose may be applied for the enhanced release of radioactivity from biomass according to recent studies in treatment of waste from papermaking (5) at an approximate price of $20 per 1,000 kg of waste.

TABLE II

Remaining Mass and Volume After Three Months of Digestion in Percent of the Initial

	Solid		
	Mass	Volume	Liquid Volume
Fresh Cress	12	2	75
Dry Cress	20	4	71
Dry Wheat	37	15	78

As shown previously, the liquid containing particles was binding the large part of strontium. If an efficient method could be managed for decantation, the liquid will mainly contain caesium. Only 50% of this caesium is under ionic form proven by strong cation exchange resins in batch experiments. After centrifugation at 11,500 g, 20% of caesium in the liquid phase is bound to particles bigger than 0.01 μm proven by membrane filtration. Therefore, it may be concluded that most of the caesium is still under organic form, bound as well to organic acids little dissociated as to very small plant particles or cell debris. Therefore, further treatment of liquids will necessitate both filtration techniques and sorption processes. Such techniques are largely described in previous Waste Management Proceedings (6, 7) and the use of inexpensive material like zeolites, for example (8), and mineral clays may be useful for this purpose. Moreover, subsequent biomineralization in aerobic conditions may transform caesium to ionic species susceptible to concentrating sorption on various matrixes. Complete biomineralization of the organic caesium containing liquid may improve the safety of final or intermediate disposal.

CONCLUSION AND PROPOSALS FOR CONTAMINATED CROP MANAGEMENT

Although the problem of contaminated crops seems to be solved only partly because of the transfer of the radioactivity to a liquid form, this option presents greater advantages for a further safe treatment of waste. Firstly, liquids are easier to transfer than biomass itself and secondly, liquids can be transported for subsequent treatment and final storage and/or disposal if distances, general geographics and risk analysis are not in contrast with safety reasons.

In anaerobic conditions, the decomposition of fresh or moisturized biomass allowed the release under liquid form of about 50% of caesium and 25% of strontium present in the internal plant. Running water washing of the remaining biomass could increase the recovery of strontium and caesium up to 70% and 80%, respectively. The remaining biomass and volume were respectively 40% and 15% of the initial for wheat and yet more efficient for cress (Table II).

Anaerobic digestion is fortunately easier to achieve and to manage in the field than an aerobic one. The technical means necessary are quite simple. A polythene sheet of e. g. 0.3 mm thickness covering the biomass pile is the main tool to supply. If correctly fixed on the pile against the wind and maybe game, the plastic sheet will prevent resuspension of contaminated dust and maintain as well anaerobiosis and the moisture necessary for the decomposition.

For liquid collection, the pile may be placed in a small excavation of the soil over a base plastic sheet. If the soil is not a clayey type, a clayey soil layer brought under the pile will filter and/or adsorb eventual leakage of liquids through the plastic sheet and prevent the contamination of the underground. Clay is the best adsorbant for radionuclides and especially caesium (8). After about two months of biodegradation, the released liquid will be pumped and transported for subsequent treatment (or only filtered through a clayey soil). The drained liquid will be replaced by running water and recovered again after about one week for further treatment.

As shown in Fig. 2, a good digestion efficiency may be expected using a liquid volume of 2 cm^3 per gram of biomass. The water volume for moisturizing, e.g., 2 Gg/km^2 of wheat, dry biomass would lead to a total volume of 4,000 m^3. According to available data (9) on caesium-137 fallout in the Chernobyl zone, deposits exceeding 0.5 TBq/km^2 covered 10,000 km^2, of which 40% were agricultural fields like wheat. In this case, the total liquid volume involved would be 4.107 m^3, considering excavations of 2 m in depth. Thus, the waste treatment area will only occupy a surface of 0.08% of the rehabilitation area. The activity of liquids and biomass would be respectively 138 MBq/m^3 and 0.27 Mgq/kg. The price of crop harvest and treatment would amount to 35,000 \$/km^2. Taking into account decantation and mineralization cost of organic matter treatment of urban liquid wastes of about 0.1 \$/m^3, the total price for the Chernobyl example would amount to about \$150 million, excluding transport, a price reasonably low for the expected efficiency of a rehabilitation method such as crop clearing and waste management.

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