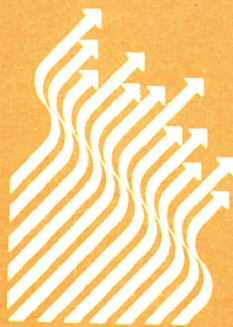




Commission of the European Communities

radiation protection

Improvement of practical countermeasures: The agricultural environment Post-Chernobyl action



Report

EUR 12554 EN

Commission of the European Communities

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Improvement of practical countermeasures: The agricultural environment Post-Chernobyl action

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Preface

The Chernobyl accident, which occurred on 26 April 1986, presented major challenges to the European Community with respect to the practical and regulatory aspects of radiation protection, public information, trade, particularly in food, and international politics. The Chernobyl accident was also a major challenge to the international scientific community which had to evaluate rapidly the radiological consequences of the accident and advise on the introduction of any countermeasures. Prior to the accident at Chernobyl, countermeasures to reduce the consequences of radioactive contamination had been conceived largely in the context of relatively small accidental releases and for application over relatively small areas. Less consideration had been given to the practical implications of applying such measures in case of a large source term and a spread over a very large area.

The Radiation Protection Research and Training Programme was influential in a number of important initiatives taken within the Community immediately after the accident. Information was collected by Community scientists and, from it, an assessment made within days of the possible consequences. This showed that the health impact on the population of the European Community was not expected to be significant. About four weeks after the accident, the Programme, together with the US Department of Energy, organised a meeting in Brussels during which the data on dispersion of radioactive material were discussed and evaluated. Several other meetings followed soon after on the transfer of radionuclides in the food chain and possible health effects. These meetings were carried out in close co-operation with the DG XI (Directorate General, Environment, Consumer Protection and Nuclear Safety) within the CEC, and, externally, with international organisations such as the International Atomic Energy Agency (IAEA) and the World Health Organisation (WHO). In addition, the Commission convoked a Committee of high-level independent scientists to assess the scientific evidence from current research in view of recent nuclear incidences, to consider the possible implications for the Basic Standards and emergency reference levels and to advise the Commission on future action in radiological protection including research. (EUR 11449 EN).

Soon after the accident, additional research requirements were identified by the Programme; these were mainly better methods to assess accident consequences and

the further improvement of off-site accident management. Several existing contracts were reoriented and new contracts were placed; however, the financial means then available within the Programme were insufficient to fund the additional research identified as necessary. A proposal for a revision of the Programme was, therefore, elaborated in 1986. It comprised 10 specific "post-Chernobyl" research actions. This revision, with an additional budget of 10 MEcu for a period of two years, was adopted by the Council of Ministers on 21 December 1987. With the help of the Management and Coordination Advisory Committee (CGC) "Radiation Protection" a number of institutes was identified to carry out the research in a co-operative manner, and the research began in the spring of 1988.

These post-Chernobyl activities have now been completed. Detailed reports on each of these studies and an additional volume containing the executive summaries of all reports are now available.

- Evaluation of data on the transfer of radionuclides in the food chain,
- Improvement of reliable long-distance atmospheric transport models,
- Radiological aspects of nuclear accident scenarios,
 - A. Real-time emergency response systems,
 - B. The RADE-AID system,
- Monitoring and surveillance in accident situations,
- Underlying data for derived emergency reference levels,
- Improvement of practical countermeasures against nuclear contamination in the agricultural environment,
- Improvement of practical countermeasures against nuclear contamination in the urban environment,
- Improvement of practical countermeasures: preventive medication,
- Treatment and biological dosimetry of exposed persons,
- Feasibility of studies on health effects due to the reactor accident at Chernobyl.

The research undertaken within the "post-Chernobyl" actions has added considerably to the understanding of the basic underlying mechanisms of the transfer of radionuclides in the environment, of the treatment of accident victims and of how the environmental consequences of accidents may be mitigated. In addition, progress has been made in the setting up environmental surveillance programmes development of predictive and decision-aiding techniques, the implementation of

which will lead to significant improvements in off-site accident management. Several new ideas and lines of theoretical and practical research have originated from the post-Chernobyl research and these have already been integrated into the ongoing Community Radiation Protection Research Programme. A further important feature which should not be overlooked, is the close and effective collaboration of many institutes in the research; this has markedly strengthened the ties between Community institutes and scientists. The outcome of all of this work is that the Community and all other countries are now better prepared and co-ordinated should a significant release of radioactivity ever occur again

Further research is continuing within the current Radiation Protection Research and Training Programme 1990-1991 on a number of the "post-Chernobyl" topics; these also form part of the proposal of the specific Programme on "Nuclear Fission Safety" 1992-1993, e.g. real-time emergency management systems, development of countermeasures in the agricultural environment, treatment of radiation accident victims, etc. Moreover, the Community Programme is currently making a significant contribution to an international evaluation, being undertaken by IAEA at the request of the Soviet Government, on the consequences in the USSR of the Chernobyl accident and of the measures being taken to ensure safe living conditions for the affected populations.

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REMEDIAL ACTIONS AGAINST NUCLEAR CONTAMINATION IN THE AGRICULTURAL ENVIRONMENT

C.E.C. POST-CHERNOBYL ACTION NO. 6

1. INTRODUCTION

The contamination of the rural areas near the site of the accident has posed problems of reclamation from which the Community could learn. The objective of the project has been to assess the usefulness of applied and existing techniques and to develop new procedures of decontamination.

A distinction has to be made between the reclamation of areas in the immediate neighbourhood of the core of the accident and of areas further from that site (up to thousands of kilometres distant). For the distinct zones established after a possible accident, a scale of countermeasure techniques are available to face the different levels of contamination.

It is quite clear that reclamation of rural areas after accidents (near and far fields) had not been given much attention in the pre-Chernobyl era. The research indeed both by Commission contractors and extraneously has been very limited so far, although information has been sought in studies on the matter and related to the research in other fields such as results of food processing studies (very scarce information) and soil-plant transfer factor studies (much more abundant).

Information produced in the USSR and related to reclamation of the site and nearby rural areas have been gathered. A number of local interventions aimed at the immobilization of the removal of contaminated soils have been carried out by the Russians and have yielded information which has been evaluated.

The consequences of nuclear accidents are however sufficiently important to justify the launching of studies aiming at establishing techniques to reduce the health and socio-economical detriment. It is emphasized that countermeasures however should not disturb normal life too much, but should on the contrary facilitate the return to normal life. They must moreover be practical and cost-effective and the end product should really be clean and acceptable for the consumer.

The main classes of countermeasures envisaged are:

- Treatment of vegetative cover present at the time of the accident.
- Chemical treatment of soil.
- Mechanical treatment of soil.
- Alternatives to "in situ" vegetation and soil treatment:
 - Selection of crops and soil uses.
 - Food processing.

2. PAST HISTORY OF REMEDIAL ACTIONS AGAINST NUCLEAR CONTAMINATION IN THE AGRICULTURAL ENVIRONMENT

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2.1 INTRODUCTION

In this chapter information has been collected regarding the different scenarios in which, an accident has happened, a nuclear test has taken place and high radioactivity contamination was produced with the consequent impact on the agricultural environment. Information about laboratory and field research on countermeasures applied to land recovery has also been collected.

The scenarios which have been considered as interesting from this point of view are listed in table 2.1 in chronological order of contamination. Their classification is as follows:

- Accidents: Windscale (UK), Palomares (Spain), Thule (Greenland), Rocky Flats Plant (USA) and Chernobyl (USSR).
- Nuclear tests: Bikini and Enewetak atolls, and the Nevada Test Site (USA).
- Others: Hiroshima, Nagasaki (Japan) and Los Alamos (USA).
- Laboratory and field research studies related with countermeasures to recover soil.

Table 2.1 List of scenarios considered

<u>Scenario</u>	<u>Start of contamination (year)</u>
Los Alamos	1942
Hiroshima	1945
Nagasaki	1945
Bikini atoll	1946
Enewetak atoll	1948
Nevada Test Site	1951
Windscale	1957
Rocky Flats Plant	1958
Palomares	1966
Thule	1968
Chernobyl	1986

For each of these scenarios the following information has been reviewed and summarized:

- Kind of contamination: Radionuclides, concentration and distribution, dose rate distribution, area involved,...
- Characterization of the area: Types of soil, types of vegetation (crops, wild plants, grass, lawns and trees).
- Countermeasures applied for:
 - Decontamination of soil:
 - Removal (depth, surface, volume, machines used).
 - Leaching.

- Decontamination of vegetation:
 - Removal (methods and machines used).
- Treatment of soils to reduce transfer of radioactivity:
 - Ploughing (depth and machines used).
 - Application of chemicals.
 - Application of fixatives.
- Alternatives of treatment:
 - Change of crops.
 - Crops management.
 - Food processing, etc.

2.2 SCENARIOS

A general description of the areas and the levels of contamination are firstly presented for each of the scenarios of interest. A summary of the countermeasures applied is, also, briefly shown later.

2.2.1 GENERAL DESCRIPTION

2.2.1.1 Los Alamos scientific laboratory

In 1942, the site of Los Alamos was selected for the development, assembly, and testing of the USA atomic bomb. An isolated site, located on the Pajarito Plateau on the eastern slope of the Jemez Mountains in north-central New Mexico, was chosen. In this area 54 buildings and adequate land were available and about 20000 hectares (49000 acres) were acquired in order to construct new facilities.

Essentially much of the mesa surface in the Los Alamos area is tuff, a soft volcanic rock.

At first, technical facilities were built up mainly on a ~16 hectares (40 acres) site known as the Main Technical Area. Between 1943 and 1945, most of the theoretical, experimental, and production work involving radioactive material essential to the development of the first atomic bomb, took place there. In early 1947 these facilities became known as the Los Alamos Scientific Laboratory (LASL), to be operated by the University of California for the US Atomic Energy Commission (AEC). After 1949, the Laboratory was expanded across Los Alamos Canyon and the Main Technical Area became known as Technical Area-1 (TA-1). Throughout the 1950's period, parts of the activities devoted to developing fission weapons were carried out in TA-1 facilities, and some of these activities continued there until 1965. Radioactive materials, including uranium, plutonium and fission products, were used at TA-1 and varying degrees of radioactive contamination of some of the buildings, waste-handling systems, and land was produced.

During the period 1943-1964, the liquid wastes generated by the LASL research activities, containing isotopes of strontium, cesium, uranium, plutonium, americium and tritium, were discharged into the Acid and Pueblo Canyons. These two are among numerous canyons cut into the Pajarito Plateau, approximately 40 km northwest of Santa Fe-New Mexico.

In 1966 the land occupied by TA-1 was given to Los Alamos Country or sold to private interests because it lay in a central area useful for the future development of the townsite and residual radioactive contamination

was not considered hazardous. Development of public facilities and commercial establishments began shortly afterwards (1).

In 1984 Acid and Pueblo Canyons were used for recreational activities. However, future residential and commercial development was conceivable (2).

2.2.1.2 Hiroshima and Nagasaki

In August 1945 the United States Army plane B-29 Enola Gay, together with two weather observation planes invaded the sky over Hiroshima, Japan, from the northeast and released the atomic bomb. The altitude of the bomb burst point was 580 ± 15 m.

Three days later, a second atomic bomb exploded in the sky over Nagasaki city. The altitude of the bomb burst point was 503 ± 10 m.

With the explosion of the atomic bombs, the epicenters instantaneously reached a maximum temperature of several million degrees centigrade and an atmospheric pressure of several 100000 bars; with the formation of a fireball, powerful heat rays and radiation were emitted in all directions within a short interval. Radiation extended not only directly from the burst point, but also from the surface of the ground-from fission fragments and the residue of neutron-induced radioactive materials (3).

2.2.1.3 Bikini atoll

After World War II, field nuclear testing first took place at Bikini atoll-Pacific ocean. During the period 1946 through 1958, this atoll was the site of approximately 23 nuclear detonations. Discrete test series were conducted in 1946, 1954, 1956 and 1958, and included both fission and fusion devices. Due to their proximity to the detonations sites, or to the variations of the weather, all of the islands of the atoll were contaminated to some extent by the radioactive fall-out (4).

The atoll consists of a number of small islands on an elliptical coral reef surrounding a lagoon with major and minor axes having dimensions of 35 and 27 km, respectively. The total land area, coral rock and coralline sands, is about 6 km^2 , and the land height generally averages 3 to 5 m above mean sea level.

2.2.1.4 Enewetak atoll

In 1947, the US Atomic Energy Commission announced that it was "establishing proving grounds in the Pacific for routine experiments and test of atomic weapons". From 1948 new nuclear operations also began at Enewetak atoll.

During the following ten years 43 detonations test took place, 3 in 1948, 4 in 1951, 2 in 1952, 1 in 1954, 11 in 1956 and 22 in 1958. No additional test were conducted at Enewetak or Bikini atolls (5).

The Enewetak atoll has about 388 square miles (1005 km^2) of lagoon and about 2.75 square miles (7.12 km^2) of dry land. The land area consists of 46 islands irregularly spread around the lagoon perimeter. Rainfall in the vicinity of Enewetak averages about 60 inches (150 cm) annually.

The soils are basically coral rock and coralline sands with minimal organic content and limited water holding capacity.

2.2.1.5 Nevada Test Site

From 1951 to 1975, the Nevada Test Site (NTS) has been used as an area for conducting nuclear detonations, nuclear rocket-engine development, nuclear medicine studies, and miscellaneous nuclear and non-nuclear experiments.

Atmospheric nuclear test were conducted periodically from 1951 to 1958, in which year a testing moratorium was implemented (6).

The NTS has an area of about 3500 km² and varies from 40-56 km in width (east-west) and from 64-88 km in length (north-south). This area consists of a large basins or flats about 900-1200 m above mean sea level surrounded by mountain ranges 1800-2100 m. The climate is variable, primarily due to altitude and the rugged terrain. Generally, the climate is referred to as continental arid. The average annual precipitation ranges from about 10 cm at 900 m altitude to around 25 cm on the plateaux.

The soil on and around the Nevada Test Site is primarily of volcanic origin. The valleys are composed of gently to moderately sloping alluvial fans and terraces. The soil is of coarse texture with low organic content and low water-holding characteristics.

2.2.1.6 Windscale

The Windscale works site of the UK Atomic Energy Authority at Sellafield, Cumberland, is primarily concerned with the production of plutonium. It consists essentially of two air-cooled, graphite moderated, natural uranium reactors together with the chemical processing plant required for the treatment of irradiated uranium metal, and the associated analytical and research laboratories.

The Windscale works site is located on a low-lying coastal strip of West Cumberland in the Northwest of England. Inland from the Works the ground rises to about 600 m. in a distance of 6,5-8 km.

The accident in October 1957 occurred during a deliberate release of stored Wigner energy from the graphite core of one of the reactors (7).

The first indication of an abnormal situation was provided by a routine measurement of air activity. The dust collection indicated an air contamination level approximately 10 times the normal level resulting from the decay products of radon and thoron. Complementary measurements made confirmed the fact that there had been a release of activity to the atmosphere.

A visual inspection through a plug hole in the charge face of one reactor revealed glowing fuel cartridges. Subsequently it was found that there were 150 channels containing uranium cartridges glowing at red heat.

2.2.1.7 Rocky Flats Plant

In Rocky Flats, Colorado, a plutonium fabrication plant for the

weapons program is operated by the Dow Chemical Company under contract to the US Atomic Energy Commission.

In 1958 an outside drum storage field was established, and drums containing plutonium-contaminated machine oils were stored there over the years. In 1959, a drum leakage was discovered in the storage area and, in 1964, evidence of drum deterioration was found and soil contamination was generated due to the oxidation of plutonium metal to PuO_2 , with an average diameter of the PuO_2 particles of 0.2μ .

During the years 1967 and 1968 the drums were removed and decontamination work started. The cleaning activities in this area added to high speed wind contributed to the spreading of radioactivity to adjacent areas (8).

The soil is very rocky containing only approximately 20% clay and organic matter. A typical soil profile in the Rocky Flats area contains three distinct horizons: 1) The dark top soil layer is usually 15 to 20 cm thick. 2) A rocky zone rich in limonite and hematite-coated minerals. This zone runs from 25 to 45 cm in depth, but is missing under the pad. 3) A layer of tan bentonite clay. This layer varies in thickness and is located at a depth of 40 to 75 cm below the surface. The total cover over the Rocky Flats area consists of this rocky alluvial material (48).

2.2.1.8 Palomares

In January 1966 an aviation accident took place over the village of Palomares in the province of Almeria, Spain, during an in-flight refueling operation. As a result of the accident a USAF bomber and its four thermonuclear bombs were lost. Two of them impacted in the earth; that produced the fragmentation and oxidation of their uranium and plutonium. These events resulted in the contamination of agricultural land (9).

The soils of Palomares, according to their petrographical composition, can be classified as lithograywacke-phyllarenite with a 50% matrix where the granular components (grave-sands) are fragments of rocks, mainly shale-type metamorphic rocks (10).

The average mineralogical composition is dominated by moscovite-illite, quartz and carbonates (39, 38.5 and 21.4% respectively) with, also, 3% of iron oxides and opaque minerals, 1.4% of chlorite and 2% of vegetable remains.

The average content of organic carbon is 0.27% and humic acids represent 47% of the total organic carbons (10). The studied soil samples were mainly composed of silt and sand. The proportion of sand ($t > 63 \mu$) predominates with about 48% against 38% of silt ($63 \mu > t > 5 \mu$) and 14% 'clay' ($5 \mu > t$).

Farming procedures in this area are typical of the Mediterranean agricultural zones of scarce yearly precipitation, about 200 l/m^2 , which require artificial means of irrigation.

2.2.1.9 Thule

In January 1968 a USA bomber carrying nuclear weapons caught fire and

crashed on the ice of the Bylot Sound, between Saunders Island and the mainland, 11 km west of Thule, Greenland (11).

At the moment of impact the conventional explosives of the four hydrogen bombs were detonated, with the result that all parts of the plane were blown apart. Around the point of impact the ice was broken to a distance of approx. 25 m, but this area immediately refroze.

The spread of plutonium into the atmosphere, to water and to the ice and snow, was due to the tremendous fire which developed.

The mean depth of the Bylot Sound is 100 m and in the impact area the depth was nearly 200 m.

2.2.1.10 Chernobyl

The Chernobyl nuclear power station is located in the eastern part of a large region known as the Byelorussian-Ukraine Woodlands, beside the river Pripyat which flows into the Dniepr.

As a result of the accident that occurred on 26 April 1986 at Unit 4 of the Chernobyl NPP, a significant quantity of the radioactive materials accumulated in the reactor during its operation escaped from the plant. It has been estimated that 100% of the noble gas radionuclides and about 3-4% of the remaining radionuclides (10-20% of the cesium, iodine and tellurium inventories) escaped from the plant (12).

The region is characterized by a relatively flat landscape with very minor slopes down to the river. About half the area is used for farming, the remainder is forested (13).

The soil cover of the contamination zone consists of light textured sandy and argillo-arenaceous sward-podzolic acid soils with a low content of nutrients, a low sorption capacity and flushing regime (wooded regions) which predetermines sufficiently intensive incorporation of radionuclides in the agricultural chains of migration (chiefly from soil to plant).

In 1986, about 49000 people lived in Pripyat town situated west of the plant's 3 km safety zone and 12500 lived at Chernobyl, the regional centre, 15 km south east of the plant. Immediate emergency response measures such as keeping people indoors, iodine prophylaxis and evacuation were carried out. About 135000 people were evacuated from a zone within a 30 km radius of the plant (13).

2.2.2 LEVELS OF CONTAMINATION

2.2.2.1 Los Alamos

- Main Technical Area (TA-1)

In 1974 a survey was undertaken to provide up-to-date information on any radioactivity in excess of background on the remaining undeveloped lands once occupied by TA-1. Fifty-five survey points were established in this area and seven additional survey points were established at locations in north-central New Mexico, 30 to 80 km from Los Alamos, to provide background

reference information. X- and gamma-radiation measurements were made at each survey point, and environmental samples were collected.

The most significant results were those for plutonium and uranium. The highest levels of plutonium contamination (224 pCi/g (8.3 Bq/g)) were found in the eastern portion of TA-1, and of uranium contamination (55 µg/g) in the western portion of TA-1, especially near buildings in which uranium operations were performed. It was found, also, others contaminants like americium, radium, cesium and beryllium, and its maximum values were 2.4 pCi/g (88.8 mBq/g), 5.2 pCi/g (192.4 mBq/g), 12.5 pCi/g (462.5 mBq/g) and 2.9 µg/g respectively. Even those maximum values were not considered high enough or widespread enough to raise the question of health or safety hazards (1).

- Acid/Pueblo Canyon site

In 1976-77 a radiological survey conducted by Los Alamos Scientific Laboratory defined the areas in Acid Canyon requiring remedial action. Four areas were contaminated in excess of background concentrations. However, only the two designated as having elevated surface activity were contaminated in excess of the remedial action criteria applicable to the Acid/Pueblo Canyon site. The pre-remedial action radiological survey data for 23 soil samples of these two areas had shown for plutonium-239 a range of contamination 0.2-163000 pCi/g, for plutonium-238 0.01-696 pCi/g, for americium-241 0.9-1200 pCi/g, for cesium-137 0.1-1/6 pCi/g and for strontium-90 0.2-229 pCi/g. The average contamination for each element was found 11787 pCi/g (436 Bq/g), 49 pCi/g (1.8 Bq/g), 129 pCi/g (4.7 Bq/g), 25 pCi/g (0.92 Bq/g) and 24 pCi/g (0.89 Bq/g) respectively (2).

2.2.2.2 Hiroshima and Nagasaki

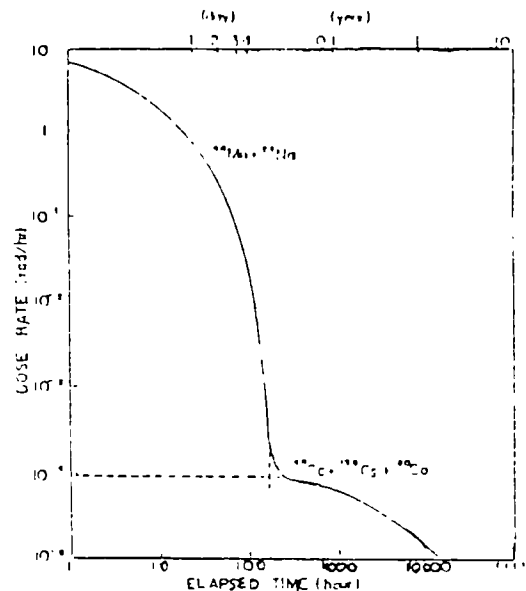
The estimated values of gamma-ray dose rate from five nuclides in soil, 1 m above ground, at various distances from the hypocenter immediately after the explosion in Hiroshima are shown in table 2.2 (14).

Table 2.2 Gamma rays exposure rate of induced radioactivity in soil as a function of distance in Hiroshima immediately after the bombing (14)

Distance from hypocenter (m)	Exposure rate				
	⁵⁴ Mn (R/hr)	²⁴ Na (R/hr)	⁴⁶ Sc (R/hr)	⁶⁰ Co (mR/hr)	¹³⁷ Cs (mR/hr)
0	4.81	2.95	0.085	0.011	4.3×10^{-4}
500	0.81	0.50	0.014	0.002	0.3×10^{-4}
1000	0.037	0.023	0.0007	0.0001	0.03×10^{-4}

Total dose rate due to these five nuclides is shown in fig. 2.1 as a function of time from immediately after the explosion. It was found that the two nuclides Na-24 and Mn-56 contributed dominantly to the gamma-ray dose from the radioactivity induced in soil (14).

Figure 2.1 Changes by time of gamma ray dose at 1 m above the ground at the hypocenter in Hiroshima (14)



The total accumulated gamma-ray dose from immediately after the explosion of the A-bombs to infinity was about 80 rads at the hypocenter in Hiroshima, and about 30 rads in Nagasaki (14).

According to the gamma ray dose at 1 m above the ground, as measured by the Japan-United States Joint Commission about two months after the explosions, a maximum 0.045 mR/h was recorded in the area 3 to 4 km to the west of the hypocenter in Hiroshima and a maximum 1 mR/h at the Nishiyama district in Nagasaki (3).

The radioactivity induced in soil at the hypocenter after the explosion in Hiroshima, was 2.99 $\mu\text{Ci/g}$ (111 KBq/g) for Mn-56, 0.89 $\mu\text{Ci/g}$ (33 KBq/g) for Na-24, 0.5 pCi/g (19 Bq/g) for Sc-46, 0.05 pCi/g (1.9 Bq/g) for Co-60 and 0.002 pCi/g (0.07 Bq/g) for Cs-134 (14).

In Nagasaki, the manganese content of soil was about two times as large as that of Hiroshima and neutron dose on the ground was about 1/3 that of Hiroshima (14).

2.2.2.3 Bikini atoll

Total exposure rates, measured in 1967, were found to vary considerably from island to island and from site to site on a given island. Levels measured in soil ranged from less than 10 $\mu\text{R/h}$ to over 500 $\mu\text{R/h}$, (external gamma radiation levels in the United States due to naturally

occurring emitters in the soil range from 0 to about 20 $\mu\text{R/h}$). On Bikini and Eneu islands the major contributor to the total exposure rate (80%) was found to be Cs-137 with minor but significant contributions from Co-60 and Sb-125. On Nam and other islands closer to blast sites Co-60 was the main contributor with important contributions from Sb-125, Rh-102m and sometimes Cs-137. Other isotopes, including Bi-207, Eu-155, Eu-152, Zn-65, Rh-106, Rh-101, Ce-144 and Am-241, were also detected occasionally (15).

On the table 2.3 are summarized the radiation levels in 1967.

Table 2.3 Summary of radiation levels on Bikini atoll in 1967
(*: No soil sample or field spectra measurement)

Island	Gamma Exposure Rates ($\mu\text{R/h}$) Range	Major Contributors
Bikini	10-120	^{137}Cs
Eneu	2- 10	^{137}Cs
Nam	10-330	^{60}Co , ^{137}Cs
Bokantuak, Iomelan,		
Rojkere, Eonjebi	3- 10	*
Aerokoj-Eneman Complex	1-570	^{60}Co , ^{125}Sb , $^{102\text{m}}\text{Rh}$
Enidrik	3-235	^{60}Co , ^{125}Sb , $^{102\text{m}}\text{Rh}$
Lukoj	60-200	^{60}Co , ^{125}Sb , $^{102\text{m}}\text{Rh}$
Jelele	60-130	*
Oroken	15- 45	*
Bokaetoktok	10- 35	*
Bokdrolul	20- 50	*
Bokbata	10- 30	^{60}Co , ^{137}Cs
Aomen-Iroij Complex	5-330	^{60}Co , ^{125}Sb

2.2.2.4 Enewetak atoll

In 1972 a radiological survey of the Enewetak atoll was carried out, and the relative amount of total fallout deposited on each island of the atoll was estimated.

The exposure rates of these islands were measured with a NaI(Tl) scintillator survey instrument held at 1 m above the surface of the location where the reading were taken. Concentration values for Sr-90, Cs-137, Pu-239, and Co-60 were obtained, for surface to 15 cm deep soil samples, in the atoll's areas where the samples were collected. In some islands this soil activity was measured separately for areas of dense or of sparse vegetation. In one case in the hot spot of Pearl island, the evaluations of radionuclides concentrations was divided into this hot spot and the remainder of the island. Table 2.4 is a summary of total fallout, of maximum exposure rates and of concentration of the radioactivity in the surface of the soils in the Enewetak atoll's islands (16).

Table 2.4 Fallout in Enewetak atoll

Island	Exposure rate		⁹⁰ Sr		Activity (pCi/g)				⁶⁰ Co	
Site name	T(H+1)*	Max ^{&}			¹³⁷ Cs		²³⁹ Pu			
(Marsallese)	(R/h)	(μR/h)	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Alice (Bokoluo)	3383	170	80	14-430	36	5.6-141	12	3.9-6.8	5.9	1.4-33
Belle (Bokombako)	3382	200	123(1)	14-670	48(1)	14-170	26(1)	7.2-130	10(1)	3.1-30
			44(2)	35-130	8.6(2)	3.3-44	11(2)	5.8-26	4.6(2)	2.4-9.6
Clara (Kirunu)	3154	100	65	13-310	26	5.6-110	22	3.5-88	6.4	0.91-20
Daisy (Louj)	3354	140	190(1)	100-380	11(1)	3.4-33	41(1)	22-98	11(1)	6.4-26
			32(2)	16-120	3.8(2)	0.86-9	15(2)	3.8-33	0.85(2)	0.37-7.4
Elna (Bokinwotme)	9533	-	46	30-220	4.2	2.7-6.4	18	13-24	0.43	0.33-0.63
Helen and Irene	5277and	8	30	5.9-570	3.2	0.22-41	11	2.4-280	5.4	0.12-520
(Bokaidrik and Boken)	6184									
Janet (Enjebi)	3501	-	44	1.6-630	16	0.57-180	8.5	0.08-170	1.9	0.02-33
Kate (Mijikadrek)	1753	-	67(1)	3.7-200	24(1)	18-37	17(1)	8.6-50	2.7(1)	1.6-5.8
			11(2)	1.6-49	4.8(2)	1.8-16	2.3(2)	0.17-14	0.46(2)	0.03-3.5
Luky (Kidrinen)	1716	-	32	10-83	11	2.2-25	7.7	2.4-22	1.5	0.26-3.8
Mary (Bokenelab)	2785	-	29	11-140	9.9	5.6-26	8.0	2.0-35	1.5	0.74-4.8
Nancy (Elle)	1251	-	36	16-110	12	6.0-28	9.1	2.3-28	1.6	0.56-5.3
Olive (Aej)	1252	-	22(1)	4.6-70	8.5(1)	3.5-28	7.7(1)	2.2-30	1.5(1)	0.65-4.1
			4.5(2)	2.0-11	0.16(2)	1.9-4.1	2.8(2)	1.9-4.1	0.11(2)	0.05-031

continue

Pearl (Lujor)	4329	-	62(3)	35-40	19(3)	7.4-55	51(3)	15-530	12(3)	3.6-70
			17(4)	3.2-61	7.6(4)	1.2-34	11(4)	0.85-100	4.1(4)	0.49-49
Percy (Taiwell)	-	-	13	3.6-73	0.94	0.12-17	3.5	1.5-23	0.47	0.08-2.9
Ruby (Eleron)	10643	110	12	7.1-63	1.4	0.71-7.2	7.3	3.0-24	0.93	0.29-16
Sally (Aomon)	1981	10	8.4	0.87-140	3.0	0.03-30	4.3	0.21-130	0.54	0.05-69
Tilda (Bijire)	774	-	27(1)	17-54	8.4(1)	3.5-20	7.6(1)	1.4-17	1.2(1)	0.61-1.9
			8.7(2)	2.2-47	1.0(2)	0.04-5.3	2.5(2)	1.1-34	0.37(2)	0.12-1.7
Ursula (Lowja)	651	-	6.8	2.0-19	1.7	0.13-7.8	1.3	0.26-7.3	0.31	0.05-1.7
Vera (Alembre)	270	-	6.3	1.1-68	20	0.03-12	2.5	0.60-25	0.30	0.02-2.2
Wilma (Billae)	294	-	3.3	0.26-13	1.3	0.13-7.2	1.1	0.1-5.3	0.12	0.01-0.7
Yvone (Runit)	62.849	-	1.7	0.09-20	0.4	0.02-3.6	3.2	0.02-50	0.64	0.01-20
Minor Southern Islands	-	1-5	0.52	0.03-3.9	0.14	0.004-1.8	0.07	0.004-1.1	0.06	0.007-63
David (Japtan)	1	5	0.55	0.08-2.6	0.39	0.03-1.0	0.05	0.004-0.23	0.03	0.009-014
Elmer										
(Madrin or Medren)	2.6	7	0.77	0.02-5.1	0.32	0.01-1.2	0.22	0.008-5.5	0.06	0.05-0.88
Fred (Enewetak)	2.6	1	0.61	0.16-1.5	0.25	0.02-2.1	0.08	0.02-0.31	0.04	0.02-0.15
Leroy (Biken)	235	8	11	1.6-34	3.2	0.5-10	0.63	0.02-2.0	0.58	0.04-5.0

(*) "Total H+1 hour exposure rate received" to estimate the relative amount of fallout deposited on each island

(&) Maximum exposure rate measured at 1 m above the surface in 1972

(1) Areas of dense vegetation

(2) Areas of sparse vegetation

(3) Hot spot of island

(4) Remainder of island

2.2.2.5 Nevada Test Site

The total area in Nevada, where the surface Pu contamination level was above 1000 pCi/cm^2 (37 Bq/cm^2), was about 300 acres (17). Only a very few acres were above 7.000 pCi/cm^2 (259 Bq/cm^2), the level where cleanup had been done in the past ((18) referenced in (17)).

Relative concentrations of ^{90}Sr , ^{137}Cs and ^{144}Ce in samples of soils collected in 1966 and 1967 at sampling sites in the Palanquin (U20K) fallout area of the NTS are shown in table 2.5 (19).

Table 2.5 Relative contribution of ^{90}Sr , ^{137}Cs and ^{144}Ce in samples of soil collected, in 1966, in the Palanquin (U29K) fallout area in Bq/g d.w. (the sites are located downwind from ground zero) (19).

RADIONUCLIDE	CONCENTRATION (Bq/g)			
	Site 4 (0.5 km)	Site 3 (1.0 km)	Site 2 (2.0 km)	Site 1 (3.0 km)
^{90}Sr	364	112	36	20
^{137}Cs	679	218	62	58
^{144}Ce	165	177	429	102

2.2.2.6 Windscale

The highest radiation level measured was 4 mR/h. The normal background radiation level in the area was about 0.01 mR/h, so that the measured value was about 400 times the natural level. It was apparent that the accident had not caused any significant external radiation exposure to individuals living in the district (7).

It was also clearly shown that the problem was basically contamination of milk by iodine-131 and that the release was not of a normal distribution of mixed fission products but had involved a preferential iodine content.

The principal fission products released were iodine-131, 20 KCi (740 TBq), cesium-137, 600 Ci (22.2 TBq), strontium-89 and -90, 80 and 9 Ci (2960 and 333 GBq). Smaller quantities of other fission products such as ruthenium-103 and -106, zirconium-95, niobium-95 and cerium-144 together with polonium-210 were also released (7).

2.2.2.7 Rocky Flats Plant

- Inside contamination

In the drum storage field, the level of Pu contamination in the soil was determined to range from 200 to 30000 kBq/m² and penetration depths of 3 to 20 cm (20).

The pad area is 113 m wide and 120 m long. Approximately 80 to 90 grams of plutonium were dispersed in $2 \times 10^7 \text{ kg}$ of soil beneath the pad (20).

It was estimated that the amount of plutonium deposited in this area was 6.3 Ci (233.1 GBq). Of that total, an estimated 1 Ci (37 GBq) of plutonium was redistributed beyond the pad area, and of that 1 Ci, 0.56 Ci (20.7 GBq) is believed to have been deposited in the 903 Lip Area (8).

- Outside contamination

Soils samples were collected to a depth of 20 cm at 33 sites extending as far as 40 miles from the Rocky Flats Plant in Colorado (belonging to the Dow Chemical Co.) (27). Deposition concentrations of Pu-239 as high as 2 Ci/km² (74 GBq/km²) were found off the plant site but these high concentrations decreased rapidly with distance.

The total Rocky Flats Pu-239 burden in the soil bounded by the 3 mCi/km² (111 MBq/km²) contour was estimated to be 4.5 Ci (166.5 GBq). The Rocky Flats Pu-239 on public and private lands not controlled by the AEC and within the 3 mCi/km² contour is estimated to be 2.6 Ci (96.2 GBq).

2.2.2.8 Palomares

The total amount of the affected area was 226.2 Ha and of that area, 2.2 Ha had a contamination level above 1200 KBq/m², 17 Ha had from 120-1200 KBq/m², 87 Ha from 12-120 KBq/m² and 120 Ha less than 12 KBq/m² (21).

2.2.2.9 Thule

Debris and fuel were spread over a drop-shaped area with the approximate size 700 x 150 m² (11). The main amounts of plutonium were confined to a limited area, and that amounts were confined into the debris.

50% of the contamination (about 25 Ci of Pu) remained on the ice surface and the rest was dissipated in the marine environment.

The maximum extra-sital plutonium deposition measured was 40 pCi/cm² ($\approx$ 14.8 KBq/m²), and the geometric mean of all samples measured was 0.4 pCi/cm². From the snow survey it was estimated that the contamination of the Thule environment outside the impact area was few curies of Pu-239 (22).

The measurements showed that from a health physics point of view, the only significant contamination was confined to the snow of the area where the fire had taken place (11).

2.2.2.10 Chernobyl

At the time of the accident, the core inventory in the reactor was about 4×10^{19} Bq (10^9 Ci). The isotopic composition of the inventory and the percentages released are shown in table 2.6. It is estimated that up to 2×10^{18} Bq (5×10^7 Ci) were released from the fuel during the accident, not counting releases of Xe and Kr. Approximately half of the material released from the reactor was deposited in the 30 km zone (13).

Table 2.6 Core inventories and total releases
at Chernobyl accident (13)

Element	Half-life (d)	Inventory ^a (Bq)	Percentage released
85-Kr	3930	3.3×10^{16}	~ 100
133-Xe	5.27	1.7×10^{18}	~ 100
131 I	8.05	1.3×10^{18}	20
132-Te	3.25	3.2×10^{17}	15
134-Cs	750	1.9×10^{17}	10
137-Cs	1.1×10^4	2.9×10^{17}	13
99-Mo	2.8	4.8×10^{18}	2.3
95-Zr	65.5	4.4×10^{18}	3.2
103-Ru	39.5	4.1×10^{18}	2.9
106-Ru	368	2.0×10^{18}	2.9
140-Ba	12.8	2.9×10^{18}	5.6
141-Ce	32.5	4.4×10^{18}	2.3
144-Ce	284	3.2×10^{18}	2.8
89-Sr	53	2.0×10^{18}	4.0
90-Sr	1.02×10^4	2.0×10^{17}	4.0
239-Np	2.35	1.4×10^{17}	3
238-Pu	3.15×10^4	1.0×10^{15}	3
239-Pu	8.9×10^6	8.5×10^{14}	3
240-Pu	2.4×10^6	1.2×10^{15}	3
241-Pu	4800	1.7×10^{17}	3
242-Cm	164	2.6×10^{16}	3

^a Decay corrected to 6 May 1986 and calculated as prescribed by the Soviet experts.

Table 2.7 shows that in the worst zone (870 km²) the surface activity was caused by 5-8.7 MCi (185-320 PBq) of contamination.

Table 2.7 Estimated surface activity in 30 km zone
of the damaged reactor (26 June 1986) (13)

Range of the activity, mR/hr	Area of zone, sq km	Activity	
		Absolute, Mcl	Relative, %
20 - >20	870	5 - 8.7	83.0
10 - 20	480	0.8-1.4	10.2
5 - 10	1100	1 - 1.7	10.8
3 - 5	2780	1.3-2.2	18.0
Total	5230	8-14	100.0

The total area of Cs-137 contamination amounts to 10000 km² for radioactive contamination in excess of 5.55×10^5 Bq/m² (15 Ci/km²); the areas with a contamination higher than 1.85×10^5 Bq/m² (5 Ci/km²) total 21000 km². Furthermore, in the north of Rovenskaya province and the south of Brestskaya province, and in zones of Kaluga and Tula, radioactive spots of a lower density, from 0.74×10^5 to 1.85×10^5 Bq/m² (2 to 5 Ci per km²), were detected, in soils characterized by an extremely low content of humus substances, with the result that the rates of cesium migration through the food chain are fairly high (23).

Because of the meteorological air mass transfer conditions prevailing, the cloud formed at the time of the accident left a radioactive trail over the area to the west and north of the plant. In the following 10 days, an intense release of radioactive gases and aerosols continued, resulting in the contamination of terrain in different directions and at considerable distances from the plant (24).

While it is difficult to assign quantitative parameters to the initial fallout pattern, some indication can be made on the basis of available data. An assumed initial angular plume of 10° extending to 60 km from Chernobyl would correspond to a segment fallout area of approx. 300 Km². If 2% of the reactor's total radioactive content i.e., one half of the total emission had been deposited uniformly over this area it would result in a deposition of some 5×10^8 Bq/m² of significant radionuclides (iodine, cesium, strontium, ruthenium, plutonium). However, if the remaining half of the total emission were uniformly deposited over the total affected area of Europe of some 10⁷ Km² this would correspond to an average deposit of the order of 10⁴ Bq/m².

In fact, the deposition was very far from uniform and on the very distant affected areas were no levels so high as to indicate the need to remove topsoils. Levels deposited were, also reduced by the radioactive decay of shorter-lived radionuclides, such as iodine-131, during atmospheric transport and after deposition. Moreover, the fraction deposited over mainland Europe outside the USSR must have been considerably less than one half because of its widespread detection beyond Europe. A fraction of one quarter has been suggested (25).

2.2.3 COUNTERMEASURES

The different countermeasures taken in each scenario are presented below. The application of some of them will be developed in the following chapter.

Los Alamos

- Removal of soil

Bikini

- Stripping of the vegetation
- Reduction of activity by physical decay
- Removal of soil

Enewetak

- Devegetation
- Ploughing
- Removal of soil

Nevada

- Vacuum cleaning
- Ploughing
- Scraping
- Fixation by oiling
- Oiling + scraping
- Wetting down with water and scraping
- Flooding with water and leaching

Windscale

- Milk consumption restriction

Rocky Flats Plant

- Fixation by gravel cover + asphalt
- Application of polyurethane foam
- Removal of soil
- Transuranides clean process (TRUclean process)
- Scrubbing and screening

Palomares

- Devegetation
- Removal of soil
- Ploughing

Thule

- Ice removal

Chernobyl

- Devegetation
- Removal of soil
- Ploughing
- Application of chemicals and fixatives

It seems clear that these decontamination measures can play a major role in reducing the external gamma radiation exposure received, particularly in regions near the contaminated areas, the inhalation of radionuclides, as a result of secondary dust formation, and the transfer of contaminants from the soil to crops and through the food chains.

These special treatments have been used and recommended according to the soil type, vegetation cover, climatic conditions, composition and type of radionuclides and other specific conditions. In these treatments could be included other decontamination methods, i.e. the changing of the traditional system of soil cultivation and the changing of harvesting and crop processing methods.

The selection of methods and technical procedures for the radioactive decontamination of the grounds is governed by a number of overriding factors (26):

- The exposure dose rates, which govern access to the scene of operations.
- The nature of the location and of the items for decontamination.
- The mechanical properties of the material forming the surfaces to be treated.
- The availability of appropriate methods of decontamination and of technical facilities for applying them.
- The experience of carrying out the work (i.e. evaluation of its economic and technical effectiveness).

2.3 APPLICATION OF THE COUNTERMEASURES

The different countermeasures are described below for each scenario where they were applied. The machinery used and the efficiency, cost and practicability are summarized where possible.

2.3.1 DEVEGETATION

The removal of vegetation with high level of contamination has been referred clearly only in the cases of Palomares and Chernobyl.

Additional experience about devegetation was obtained from the preparation for agricultural redevelopment of affected areas with lower level of contamination (i.e. Bikini atoll) or from the preparation by lane cutting of these areas for radiological survey and from the elaboration of specific devegetation experiments (case of Enewetak atoll).

2.3.1.1 Bikini atoll

The cleanup operations on this atoll included the stripping of the vegetation in Bikini and Eneu islands to permit planting of coconuts, pandanus and breadfruit (4).

- Bikini and Eneu islands

The islands were prepared for agricultural redevelopment by cutting parallel strips through the vegetation along the length of each island. The strips, out on 56-foot (17.1 m) centers, were approximately 20 feet (6.1 m) wide. The vegetative cover was knocked down and left in place to provide additional organic matter for the soil which did not provide a real decontamination. The strips were surveyed and background gamma radiation levels recorded at 250 feet (76.2 m) intervals along their length (4).

- Efficiency

Experience obtained during the clearing operation indicates that total removal of the vegetation cover and turning of the soil as occurred during the grading of the perimeter and cross island roads results in a rapid reduction of the measured exposure rate.

2.3.1.2 Enewetak atoll

Island Janet was selected as the location for devegetation experiments. The method selected concerning an area measuring about 1000 x 1000 feet² (304.8 x 304.8 m²) consisted of dragging a 200-foot (61 m) anchor chain across the brush. Two bulldozers, each with an end of the chain attached, drove in parallel across the terrain, keeping the chain just slightly slack. This system worked well in areas with only moderate vegetation. In specially dense growth, the chain would only partially knock the brush down, so a second pass was required in the opposite direction to the first pass. The brush was, at this point, still a tangled mass which the vehicle could not traverse. The next action was to push the brush into windrows about 150 meters apart. The bulldozer operations maintained the

bulldozer blade about 6 inches (15 cm) above ground level, but a substantial volume of dirt was still pushed into the windrows. The windrows remained in place until near the end of the next dry season, when they were eventually all burned with the aid of liberal doses of diesel oil (5).

- The lane cutting experience

Early devegetation experiments on Janet clearly indicated that a more expeditious method would have to be found for preparing an island for the coarse-grid IMP survey (IMP was the manufacturer's name for the tracked vehicle used to house the in situ measurement equipment and this term was often used to refer to the entire system). Total removal of brush consumed too many man and machine resources, was too slow, introduced too much soil disturbance, and was not necessary for measurement of Am-241 gamma emissions. The last areas on Janet to be prepared for IMP access were not heavily vegetated so the bulldozer operator was instructed to push aside only that brush which interfered with line-of-sight surveying and staking by the Army engineers.

Experiments continued from island to island as new combinations of brush density and soil hardness were encountered. By the time lane clearing was completed on the major islands, the methodology had evolved to eliminate setting the blade down into the soil. The new method was found to set the blade about four to six inches (10-15 cm) above the soil surface. This was found to be suitable for knocking down the larger trees and breaking off the smaller brush. Occasionally some trees would be uprooted and the stump and roots would have to be pushed aside but, in usual conditions, a lane could be cleared with minimal soil disturbance. The bulldozer operator had only to try not to leave material in the lane that could protrude up into the engine compartment of a passing IMP, or that would be too rough for the low-clearance IMP to negotiate (5).

- Practicability

The vegetation cover had to be removed, before soil removal could follow, and several methods for accomplishing this were tested and evaluated.

The most rapid technique was pushing vegetation into windrows with a bulldozer, as done on Janet island, but this method mixed and spread the surface soil so that high levels of surface contamination could be spread over a larger area than initially existed. Also, the windrow would contain a substantial volume of contaminated soil which could not easily be separated from the vegetation.

The second method utilized a front loader with what is called a four-in-one bucket*. This machine was initially tried and determined to be unsuccessful because it did not remove many roots and bush stumps. After realizing the drawbacks of windrowing by bulldozer, the bucket loader was reevaluated. Small bushes or brush could be effectively removed with minimal soil disturbance by lowering the bucket to six inches above ground and making

* Four separate hydraulic controls governed all possible motions of the bucket, including the ability to clamp items between longitudinal halves of the split bucket.

a forward pass up to 50 feet (15.2 m) long. For larger bushes, the bucket was clamped over the bush and the whole bush plucked from the soil and carried to the brush pile. The latter technique created the least disturbance of surface soil. A road grader with scarifier teeth was determined to be least satisfactory as a means of removing stumps and roots.

2.3.1.3 Palomares

Harvesting of contaminated crops had been made traditionally. Machetes were used to cut tomatoes and other crops, as well as sickles, scythes and hoes. As tomato crops required cane poles for their growth, three-limb shredders were requested. Cane poles were pulled from the ground, shredded, and the remains loaded into the trucks for delivery to the storage site.

A burning area was used to destroy the vegetation removed with contamination under 400 cpm*, and cane poles. A total of 3700 truckloads of vegetation (2.5 tons each) were hauled from the area and destroyed (table 2.8). In a normal day operation, 140 truckloads were moved either to the storage location or the burning area (27).

In site 2 of Palomares, about 400 truckloads hauled to the burial area, and 700 truckloads of crops were hauled to the destruction site for burning.

Removal of crops at site 3 was made during five weeks, with 2815 truckloads being burned and about 165 going to the disposal trench.

Table 2.8 Final disposition of the contaminated vegetation in Palomares(27)

a. removed, mulched, stored (400 cpm* or higher on vegetation)	400 yd ³ (305.8 m ³)
b. burned and ashes stored (less than 400 cpm* on vegetation)	3700 truckloads (2.5 tons each)
c. all removed and buried from areas where soil was above 700 cpm	285 acres (17 Ha)

* : 13000 cpm correspond to 1000 $\mu\text{g}/\text{m}^2$ of Pu ($64 \mu\text{Ci}/\text{m}^2$ or $2.4 \text{ MBq}/\text{m}^2$).

2.3.1.4 Chernobyl

Agricultural harvesting of crops was carried out in the exclusion zone and the strict control zone using special protection measures worked out by the State Agricultural Programme of the USSR and Ukraine SSR and the USSR Ministry of Health. After the harvesting, lime, mineral fertilizers and sorbents were added to the soil to increase fertility and reduce entry of radionuclides into the new crops.

The special requirements for harvesting were as follows:

- Mechanical cultivation was kept to a minimum to reduce dust formation.
- The grain and industrial crops were harvested by direct combine harvesting.
- Depending on the contamination level of the crops, the grains were used (after being stored) for food purposes, fodder, seed and industrial processing.

Research has shown the forests act as accumulators of radioactive substances, first in the crown and then in the forest litter. The radionuclides fixed in the litter will be excluded for a long time from radiation chains. At present, the majority of experts believe that the best way to deal with contaminated forests is to increase the fire prevention service.

An area of pine trees adjacent the site received a lethal dose of radiation and the trees have now been removed (28).

Although most trees were left to let nature do the decontamination, some trees near the site were cut down, bulldozed into deep trenches and covered with soil. The bottoms of the trenches were above the water table.

2.3.1.5 Field researches and laboratory tests

- Removal of crops and mulches

The presence of a crop would affect the choice of treatments for a contaminated land. Removal of the crop would partly decontaminate a land area and, in some cases, crops might have to be removed before other more effective treatments could be carried out.

A comparison of methods for removing contaminated crops or mulches from land is presented in table 2.9. The effectiveness of the treatment, the effort required for treatment and for disposal of the contaminated material, and the productivity of treated land are included (29).

With one exception, none of the methods removed more than 75% of simulated fallout from a contaminated area. In view of the rather poor removal of radioactivity, crop removal would probably be used only as a necessary preliminary task to some latter soil treatment that would be more effective.

Roots that could not be cut might decrease the effectiveness of scraping.

Crop removal requires considerable time (less than 5 acres per hour).

Note: The problem of contaminated plant material disposal is very important and has received little attention.

Table 2.9 Comparison of methods for removing contaminated crops or mulches from land (29)

Type of vegetation	Implement	Removal of radioactivity ¹	Effort required—	
			For removal ²	For disposal ³
Soybeans, 12" high	Mower	Poor	Poor	Fair
Soybeans, 12" high	Full harvester	Poor	Fair	Good
Soybeans, full growth	Full harvester	Poor	Poor to fair	Good
Soybeans, full growth	Forage harvester	Poor	Poor to fair	Good
Soybeans, mature	Combine, straw removed	Poor	Poor	Fair
Fescue-clover meadow	Forage harvester	Poor	Poor to fair	Good
Sudan grass, 12" high	Mower	Poor	Poor	Fair
Sudan grass, 12" high	Full harvester	Poor	Fair	Good
Rye, full growth	Mow, rake and bale	Poor	Poor	Good
Rye, full growth	Forage harvester	Poor	Poor to fair	Good
Rye, mature	Combine, straw removed	Poor	Poor	Fair
Wheat, mature	Combine, straw removed	Poor	Poor	Fair
Corn, full growth	Forage harvester	Poor	Poor	Fair
Mulch, 5 tons wheat straw/acre	Side delivery rake	Good	Poor	Fair
Mulch, 5 tons bermudagrass hay/acre	Rake and bale	Poor	Poor	Good

¹ Rating of removal of radioactivity: Good—>95 percent removal.
Fair—75 to 95 percent removal.
Poor—<75 percent removal.

² Rating of removal effort: Good—>5 acres per hour.
Fair—1 to 5 acres per hour.
Poor—<1 acre per hour.

³ Rating of disposal effort: Good—additional loading and hauling effort minimal.
Fair—considerable effort in loading and hauling.
Poor—very great loading and hauling effort.

2.3.2 REMOVING OF SOILS

2.3.2.1 Los Alamos

— Criteria

Main Technical Area (TA-1)

The plan for decontamination of TA-1 site had one principal objective: Any contamination found by the exploratory efforts was to be removed to the lowest practicable levels. Contaminated surface and subsurface soil was removed to ensure that any doses from remaining contamination were "as low as reasonably achievable" (ALARA) levels. The radiation exposure rate criteria were based on the limits: 500 mrem/yr to the maximum individual and 170 mrem/yr to a suitable sample of the exposed population for the whole body, gonads, or bone marrow, and three times those values for other organs (30).

Acid/Pueblo Canyon site

Remedial action criteria applicable to the Acid/Pueblo Canyon site were on the base of external exposure rates and radionuclide concentrations in soil. The radiation exposure rate criterion was based on an annual limit

for population exposures of 170 mR. For control purposes, an exposure rate of 0.02 mR/h (20 µR/h) above background was used. Background exposure rates in the Los Alamos area are 9.4 - 17.4 µR/h.

Soil criteria for food cultivation/ingestion and resuspension/inhalation pathways were also considered. The second pathway was the more realistic basis for evaluation in the case of Acid/Pueblo Canyon, since the terrain on and near the remedial action site, is unsuitable for cultivation. Table 2.10 shows the radionuclide concentrations in soil cleanup criteria for remedial action at this site (2).

Table 2.10 Soil cleanup criteria for remedial action
at Acid/Pueblo Canyon (2)

<u>Radionuclide</u>	<u>Criteria* (pCi/g)</u>	
	<u>Food Cultivation/ Ingestion</u>	<u>Resuspension/ Inhalation</u>
Strontium-90	100	2x10 ⁶
Cesium-137	80	7x10 ⁶
Plutonium-238	100	7600
Plutonium-239	100	7600
Americium-241	20	----
Uranium (natural)	40**	2200
Radium-226	5**	7000

* Criteria are applied as average concentration per 100 m² areas.

** After extensive health effects studies, the limit for uranium (natural) was increased to 75 pCi/g in November 1983. Based on these and other studies, the limit for radium-226 was also modified in November 1983 to provide for 5 pCi/g in the first 15-cm soil layer and 15 pCi/g in successively deeper 15-cm layers.

- Soil removing methods and machinery

Main Technical Area (TA-1)

Essentially all decontamination work involved the removal of soil or tuff (soft volcanic rock). Some debris, including pipes, clay-tile sewer lines, septic tanks, concrete, and various building material, were also removed, but its volume was insignificant in comparison to that of the soil and rock.

Small isolated spots of contamination or spots with high concentrations were carefully handshoveled into plastic bags to minimize the spread of contamination. Contaminated soil removed by backhoe was loaded directly into a dump truck.

Throughout all excavation and loading operations that might result in suspended dust, water sprays were used extensively to wet the exposed earth and preclude airborne contamination.

All contaminated material was taken to a controlled solid radioactive wastes disposal area, which accommodates the routine contaminated wastes from laboratory operations.

Dumper trucks hauled contaminated or potentially contaminated material to the laboratory's solid radioactive waste disposal site ~12.5 km away. The truck-beds were lined with plastic sheets to prevent loss of material in transit. For material from uranium-contaminated areas, only the tailgate was lined; for material from plutonium-contaminated areas, the entire truck bed was lined with plastic sheeting large enough to fold over the load. All truckloads were covered with canvas tarpaulins which were tied to prevent the dirt from blowing. As a loaded truck left the work area, the tires and the ledges on the truck-bed were phoswich-monitored to assure that contamination was not tracked out. A load ticket carried by each driver gave the nature of the load's contamination to ensure proper disposition at the disposal site. A health physics monitor in a second vehicle having radio communication with the truck driver escorted any truck known to be carrying appreciably contaminated material to the disposal site (1).

Acid/Pueblo Canyon site

Contaminated material was excavated from 15 to 20 cm lifted by a backhoe and loaded directly into 18-yd³ capacity dump trucks lined with reinforced plastic. Excavation was started at the point farthest from the loading point so that contaminated material was not moved over non-contaminated areas. When it was necessary to load over a non-contaminated area, that area was covered with plastic, which was rolled up and disposed of at the end of operation. A water truck was maintained at the site during excavation so that the excavation area could be wetted to control dust.

Hot spot excavation at the former vehicle decontaminations facility was performed manually with spades and shovels. Contaminated earth was loaded into 55-gal (208 l) drums that were hoisted into dump trucks by an 18-ton hydraulic crane.

All contaminated materials were disposed of the LASL Radioactive Waste Disposal Area G (TA-54).

- Level of decontamination effort

Main Technical Area (TA-1)

The intensive exploratory efforts and the decontamination operations at AT-1 lasted ~1 yr and ~15000 m³ of contaminated and potentially contaminated material were removed to LASL's solid radioactive waste disposal site.

Acid/Pueblo Canyon site

A total of 390 yd³ (298 m³) of contaminated material was excavated in implementing this remedial action (2).

- Efficiency

Main Technical Area (TA-1)

At TA-1 area, the maximum post remedial action contamination was 120 pCi/g (4.44 Bq/g) of gross-alpha activity; this indicates that spots with activity > 1000 pCi/g (37 Bq/g) were removed along with about 90% of their contaminants. This area was considered to be decontaminated to ALARA levels.

Acid/Pueblo Canyon site

In this site, the average concentration in soil in the remedial actions area was 36 pCi/g (1.33 Bq/g) of plutonium-239.

Plutonium-238 concentrations over the remedial action area were less than 2 pCi/g (0.07 Bq/g) or less than 2% of the Pu-238 decontamination criterion (100 pCi/g or 3.7 Bq/g for food cultivation/ingestion pathway, see table 2.3.3).

Post-remedial action external exposure rate near the untreated outfall was 17 μ R/h compared to the Los Alamos area average exposure rate, background, 9.4 to 17.4 μ R/h (2).

- Cost

Excavation cost at TA-1 operations was estimated \$50/m³. The total cost of the TA-1 project (1975-1976) was 769 Kilodollars.

The total cost of the remedial actions at Acid/Pueblo Canyon was K\$1038 (1981-1983). Extensive radiological characterization and subsequent engineering analysis were the major cost contributors. However, the cost for disposal of the 390 yd³ (298 m³) at the LASL site was quite reasonable: \$19000.

2.3.2.2 Bikini atoll

All debris or artifacts having little or no useful value were removed.

Although large amounts of debris were found in this island (from testing program and World War II), with one exception, none was radioactive. One pile of rotting paper scraps contaminated primarily with Cs-137 was located northwest of center on the lagoon side of the island. This material, which showed a contact reading of approximately 200 μ R/h, was loaded into 55-gallon (208 l) drums and disposed of in the ocean south of Eneu.

Several instances of high levels, greater than 200 μ R/h, due to soil contamination were encountered. It was the consensus that attempting to reduce these levels by removing the top layer of soil would destroy the limited agricultural capability of the area, therefore, most such areas were left essentially undisturbed (4).

- Efficiency

After the clean-up operations the Bikini island was divided into essentially three domains (the range of radiation levels before the clean-up was 10-120 $\mu\text{R/h}$):

1. Beach area: Uniformly background of approximately 10 $\mu\text{R/h}$.
2. Village area: Located along lagoon side of the island. Two sets of data were obtained:
 - a) From lagoon road to approximately 250 feet (76.2 m) inland the mean background was 52 $\mu\text{R/h}$.
 - b) From lagoon road to beach-35 $\mu\text{R/h}$.Considering these two areas together the mean background would be 44 $\mu\text{R/h}$.
3. Interior: Used for agriculture. This was considered to include the area within the perimeter road, excluding the village area. The mean background was 86 $\mu\text{R/h}$.

In Eneu island, although an exposure rate of 50 $\mu\text{R/h}$ was measured at one depressed location, filling of this "borrow pit" area this level was reduced to approximately 10 $\mu\text{R/h}$.

2.3.2.3 Enewetak atoll

Five islands, Janet, Pearl, Sally, Ivonne and Irene, were the most severely affected by nuclear testing operations and are the islands which required soil removal in the cleanup (5).

- Criteria

The final criteria for surface soil clean-up were:

1. Clean all 0.5 hectare areas on food gathering islands that exceed 160 pCi/g of Pu (5.92 Bq/g).
2. Clean all 0.5 hectare areas on agricultural islands that exceed 80 pCi/g of Pu (2.96 Bq/g).
3. Clean all 0.25 hectare areas on village islands that exceed 40 pCi/g of Pu (1.48 Bq/g).
4. Transuranic activity in any 5 cm depth interval below the surface shall not exceed 160 pCi/g of Pu (5.92 Bq/g) when averaged over 1/16 hectare (5).

- Machinery

Several different combination of machines were used for soil removal: Road graders, bucket loaders, bulldozers, different-sized dump trucks and water craft for bulk hauling of soils.

- Efficiency

During the clean-up of Enewetak atoll plus than 30 Ha (70000 m³ of soil) excised and approximately 14 Ci of TRU activity removed (table 2.11).

Table 2.11 Volume and TRU Activity of soil excised during the radiological clean-up of Enewetak atoll (5)

Island	Soil Volume (m ³)	TRU Activity removed		Total removed area	
		(Ci)	(GBq)	(Ha)	(%)
Janet	40525	2.6	96.2	15.5	13.1
Pearl	11415	1.7	62.9	9.7	44.7
Sally	8100	1.3	48.1	1.8	4.5
Yvonne	8210	7.2	266.4	5.0	-
Irene	3775	1.0	37.0	0.6	3.3
Total	72025	13.8	510.6	32.6	

- Practicability

Several different combinations of machines were tested and evaluated for effectiveness and practicability at soil removal. It was quickly determined that:

- The road grader was not effective.
- The bucket loader, with the bucket down and closed, could remove about 50 to 60 cubic yards (38 to 46 m³) of soil per hour, taking a six-inch (15.2 cm) "lift" or cut.
- The bulldozer, when operated in its lowest gear, made acceptable six-inch cuts when the length of push was no more than 50 feet (15.2 m). Each successive lateral pass had only 10 to 20 percent of the blade width in new soil. The rest of the blade was used to accumulate pushed up material (Soil began to spill off the open end at about 50 feet (15.2 m)). The bulldozer could windrow about 180 to 200 cubic yards (137.5 to 152.7 m³) of soil per hour. The bucket loader would then be used to load the windrowed material into a dump truck.

2.3.2.4 Nevada Test Site

- Criteria

It was requested by the Nevada Applied Ecology Group to initially consider three definitions of decontamination: removal of surface soil (0-15 cm) containing greater than 1) 160 pCi (5.92 Bq) total transuranics per gram of soil, 2) 80 pCi/g (2.96 Bq/g) and 3) 40 pCi/g (1.48 Bq/g). These limits were chosen because were used in the Enewetak atoll cleanup operation completed before (31).

- Cleanup And Treatment (CAT) test

The US Department of Energy (DOE) proposed an investigation of a vacuum method of soil removal. This investigation was conducted by the Reynolds Electrical and Engineering Co. on the Nevada Test Site (32).

Soil was removed from the test area to an average depth of 6.4 cm. An estimated 19.4 m^3 of contaminated soil was transported for disposal. Due to bulking of the material during vacuum removal and stock-piling, a 20% bulking factor was used to estimate the actual volume of material removed from the site. Approximately 161.9 m^3 of soil was removed from the 2452.6 m^2 test area.

Efficiency

The CAT test was a rigorous test of the vacuum method of soil removal and the two $12 \times 10^2 \text{ m}^2$ plots were decontaminated to less than 10 pCi/g (370 Bq/Kg) of Am-241 contamination in surface soil. In fact, in contaminated areas with 40-131 pCi/g of Am-231 and 230-748 pCi/g of Pu-239,240, with the application of the CAT test final measurements of 1.5-9 pCi/g for Am-241 and 8.5-54 pCi/g for Pu-239,240 were obtained.

Practicability

Where contamination is present in only the surface 0 to 2.5 cm of soil, the vacuum method of soil removal presents an advantage over conventional earthmoving techniques. Since the VAC-ALL is capable of removing less than 5 cm of soil per pass, the total volume of collected material can be reduced greatly in comparison with conventional earthmoving techniques.

The vacuum soil removal technique facilitates removal of contaminated surface soil without disturbance of the vegetation root systems. The vacuum method, also, allows for selective removal of contaminated soil, and exclusion of rocks greater than 7.6 cm in diameter from collected material.

- Pahute Mesa's decontamination experience

A spill of radioactively contaminated mud occurred at the U20aa postshot drilling site on Pahute Mesa at the NTS. Cleanup operations commenced in 1976, using front end loading, vacuuming, shoveling and bagging for soil removal, and flushing with water for rocky surfaces decontamination (33 and 34).

The contaminated mud flow was approximately 2495 ft (760 m) long with variable width from about 197 ft (60 m) on the drill pad and fan-shaped area to less than 3 ft (0.9 m) at several canyon locations.

Dose rates at contact varied from 5 to 200 mrad/h. The cleanup criterion was 1 mrad/h at contact, which was the background level in the general vicinity. Samples of radioactive mud were analyzed by gamma spectroscopy and the only radionuclides present were ruthenium-103 and ruthenium-106/rhodium-106. At the time of release, about 86% of the activity was due to Ru-103 and 14% due to Ru-106/Rh-106. The mud was also found to be relatively insoluble in water.

Approximately 900 man days were expended on this cleanup and 1976 m^3 of contaminated dirt were removed. A similar amount of clean dirt was transported from about two miles away to cover the crater burial site, mud stump and areas containing residual radiation above 1 mrem/h (10^{-5} Sv/h) at contact. It is estimated that 900 mCi (33.3 GBq) of contaminant remained six months after the spill.

Machinery

In clean-up operation, front-end loaders, road scrapers, dump trucks, water trucks with hose, a 10 m³ vacuum cleaner, shovels and bags were used.

Efficiency and practicability

The effectiveness, as measured by the fraction of radioactivity remaining (FR), ranged from 10⁻¹ to 10⁻³, depending upon the method used and the type of terrain.

Front-end loading dirt with an FR of about 10⁻² (table 2.12). Shoveling and bagging achieved FRs of 10⁻² in locations of fine grain, compact dirt. Vacuuming was very effective in flat areas with fine grain compact dirt achieving FRs down to 10⁻³, but was a very slow process compared to front-end loading.

Flushing with water was the method chosen for rocky surfaces and was the primary method of decontamination on area 4. FRs down to 10⁻³ were achieved on smooth surfaces and about 10⁻¹ in cracks.

Table 2.12 shows the positive and negative points of decontamination methods of terrain. The effectiveness of these methods and the type of terrain, where these were applied, are also shown.

Table 2.12 Evaluation of soil decontamination methods at NTS

TERRAIN	DECONTAMINATION METHOD	FRACTION REMAINING (FR)	METHOD OF EVALUATION
Flat compact dirt, fine grain	Front end loader	10 ⁻¹	Positive: Rapid, good decon factor. Negative: Inexperienced driver can recontaminate clean areas.
	Vacuum	10 ⁻³	Positive: High decon factor, good where subsurface boulders are present, no experienced operators required. Negative: Slow compared to FCL.
	Shovel & bags	10 ⁻²	Positive: Good in locations inaccessible to heavy equipment. Negative: Slow, considerable care must be taken not to recontaminate cleaned areas (experienced personnel desirable).
Flat ground, coarse grain or gravel-like	Front end loader	10 ⁻¹ to 10 ⁻²	Positive: Moves large amounts of dirt. Negative: Inexperienced driver can recontaminate clean areas.
	Vacuum	Not attempted	
	Shovel & bags	10 ⁻¹	Positive: Can be used in locations inaccessible to heavy equipment. Negative: Impractical in locations where deep penetration of contaminant has occurred.
Rocky Canyon (upper drainage bed)	Vacuum	Not attempted	Positive: Can be used in soil collection areas inaccessible to FCL by adding extension hose. Negative: Most rocky areas inaccessible to vacuum even with hose extension.
	Shovel & bags	10 ⁻² (fine grain soil)	Positive: Good in locations where contaminated soil has collected. Negative: Usually too many rocks.
	Flushing with water hose or scrubbing with broom/brushing	10 ⁻³ (rocky surfaces) 10 ⁻¹ (cracks)	Positive: Good in areas where contamination is hidden under rocks or exposed on rocky surfaces. Negative: Care must be taken so contamination is not spread further; e.g., constructing a dam.

2.3.2.5 Rocky Flats Plant

The removal of soils in Rocky Flats Plant was made in the 903 Lip Area (8). The maximum plutonium contamination was 5690 Bq/g in the top 5 cm of soil. All soil that exceeded 250 counts per minute was removed.

- Machinery

During the summer of 1976, the contaminated soil was hand-excavated and placed in plastic bags and monitored. Then they were placed in plastic lined 4'x4'x7' boxes for shipment offsite, until contamination levels in area were below 250 counts per minute (detectability limit of a FIDLER).

During the summer 1978 a new technique with mechanized equipment was used, involving a front end loader alone or in conjunction with road maintainer and/or dozer. Hand digging was done in any area where mechanized equipment could not be used.

Prior to starting soil removal, the area to be excavated was premoistened by sprinkler system for three days. All contaminated soil was loaded into plastic-lined 2'x4'x7' plywood boxes for offsite shipment by truck to NTS storage.

- Efficiency

Approximately 0.5 Ci (18.5 GBq) of plutonium was removed from 903 Lip Area for two summers. This quantity is based on an average concentration of 1200 Bq/g of plutonium contamination in soil and a density of 1 g/cm³.

- Cost

In the first summer (1976), using the hand digging method, 35 boxes of contaminated soil were removed at an approximate cost of \$44000 or \$1257 per box.

Use of mechanized equipment during the second summer (1978), the cost of the 1448 boxes of contaminated soil was \$410000 or \$283 per box.

2.3.2.6 Palomares

A 10 cm deep of top layer soil in areas where surface contamination was greater than 1200 kBq/m², that was 2.2 Ha (see page 14) was removed. It was replaced by well-fertilized earth from noncontaminated zones. The soil removed was considered as radioactive residue.

- Criteria

Remedial actions were taken as follows:

1. Removal of surface soil (0-10 cm) where alpha surface contamination was more than 1200 kBq/m².
2. Wet down, ploughed to a 30 cm depth, harrowed and mixed all arable land with levels below 1200 kBq/m².
3. Removal of surface soil by hand tools on rocky hillsides where surface alpha contamination was more than 120 kBq/m².

- Methods and machinery

Soil removal was accomplished by the use of road graders, where possible, with it first being moved into windrows, and then into piles and finally loaded into trucks. Where graders could not be used, as in the isolated hilly area around Site 2, the work had to be done by hand.

When scraping left small hot spots, ploughing and/or hand removing was necessary. For low contamination, scarifying of the soil, with minimum turnover, dropped the count to acceptable limit. This minimum movement of surface area was primarily important in Site 2, where it was feared that major movement of top soil in the fragile area create a dust bowl.

Because of the extremely dry climate of Palomares, the deposits were at first fixed with mineral oil in and around the small craters where the contamination was greatest, and by abundant watering in other places where contamination was lower.

The equipment used was:

- 11 dumper trucks
- 3 road graders
- 2 bulldozers
- 2 front end loaders (2cubic yards buckets);
- 5 gang ploughs and
- 16 water distributor trucks were used to reduce and avoid the resuspension (35).

For the final deposit of the radioactive residues it was thought to dig pits and to bury them. The site was chosen in zone 2.3. The pit was a trench-silo type with an approximate 1000 cubic yard (765.6 m³). At the end it was decided to send, the waste placed in a 55-gallon (208 l) barrels, to USA.

- Cost

The costs could be evaluated on bases that 1088 cubic yards for 2.2 Ha (5.5 acres) were removed and the total waste (soil and vegetation) was placed in 55 gallon (208 l) barrels, 5300 were needed and sent to Savannah River Facility, Aiken, South Carolina, USA (36).

- Practicability

There were some problems for soil removal using road graders in the isolated hilly area around Site 2, due to slope and rocks, and a hand soil removal was then made.

2.3.2.7 Chernobyl

After harvesting, the surface layer of turf was removed in certain areas, especially near the damaged facility. In some cases the turf was consolidated using a latex emulsion (SKS-65 gp).

A layer of soil 5-10 cm thick was removed and loaded into drums which were stored in the solid waste storage vault of the fifth unit. This was the major decontamination method used for the site. Where required, concrete

slabs or clean soil were placed over residual hot spots. Concrete slabs and earth were covered by a film-forming material (13).

During the decontamination of the urban areas, after the washing of the buildings, the radioactive contamination on the earth increased about 2-2.5 times. This earth was removed with bulldozers and sent for disposal.

- Methods and machinery

Essentially, the cleanup in the vicinity of the reactor consisted of removing the contaminated surface layer of soil and at selected sites suctioning up dust or covering the source with fill or concrete.

This method was widely applied, by mechanical means. The most effective procedure was the scraping of the contaminated soil by bulldozers with biological shielding and remote control and, also, by other road building machinery (loaders, scrapers, graders). However, the bulk of the work was carried out by civil-engineering ground-clearing machines which, fitted with grab attachments located on telescopic booms, were able not only to collect but also to load soil into containers and place the latter onto transport vehicles.

The method of collection and removal of contaminated soil was the basic procedure in all cases of decontaminating "soft" features of the ground relief, i.e. within the perimeter of the power station and in the town of Pripyat, where all the ground except roads and pavements was cleaned in this way (26).

Vacuum cleaner equipment was used for collecting dust. The type of such equipment available extended over a wide range, from heavy aerodrome machines to hand vacuum cleaners. Adaptation to the new purpose consisted of replacing the existing filters with special ones possessing heightened absorption capacity for active particles.

Remotely operated vehicles used

A total of over 50 units of remotely controlled vehicles from the USSR, the Federal Republic of Germany, Finland, Poland, Japan and other countries were used during the cleanup. All machines were found to perform satisfactorily and reliably, although most have some shortcomings (28).

The main operating difficulties with the remotely controlled devices during the Chernobyl cleanup include:

- a) Inadequate reliability of both the mechanical parts and control systems. The control systems failed most frequently as a result of high radiation doses.
- b) Failure of some vehicles to overcome obstacles.
- c) Tangling of the control cable with obstacles or even with machine. Cable stowing devices improved the performance.
- d) Inability to decontaminate a vehicle quickly and thoroughly after it had worked in the active zone.
- e) Lack of modular components which can be replaced quickly when failures occur.

A general comment which applies to all the remote controlled road-building types of machine is that the operator did not have a satisfactory view and little feel for the loading. As a result, nearly all the earth decontamination work was done using biologically protected machines.

A big problem with shielded vehicles was that the weight of the shielding reduced the stability, manoeuvrability and easy control of machines (28).

Transportation and disposal of contaminated wastes

The most highly contaminated earth and rubble from the clean up of the Chernobyl site was loaded into drums and sent for storage in the solid waste storage vault of Unit 5. These drums will eventually be sent to a normal disposal site.

The large volumes of less active material on the site and in areas close to the site were bulldozed into piles using shielded and/or non-shielded machines, loaded into trucks and transported to several storage/disposal areas which had been established near the nuclear power plant site. Most of the waste was transported to the storage/disposal sites in trucks.

Much of the low level radioactive waste was placed into specially dug pits which were lined with clay or other impermeable material. The waste was placed into a mound which continued above the normal ground level. The mounds were covered with a layer of clay and a layer of soil and then seeded to produce a vegetative cover. The seepage from these sites is being monitored (28).

2.3.2.8 Thule: Ice removal

The contaminated snow, approx. 12000 m³, was removed and stored in a large number of 30 m³ steel fuel containers. The radioactive snow (water) was sent to the USA. The cleanup operation itself was carried out by US military personnel which at a time comprised more than 700 people. At a later stage this activity was supplemented by Danish civil technicians and workers (11).

The cleaning up of the crash site was very efficient (> 95% of the contaminated ice was removed) and it was estimated that only approx. 25 Ci ($\pm$ 50%) were dissipated into the marine environment (22,37).

2.3.2.9 Field researches and laboratory tests

A comparison of methods for removing contaminated surface soils in warm and cold weather is given in tables 2.13 and 2.14 respectively (29).

The equipment ranges from sweepers, which would remove a minimal thickness of soil, to heavy earth-moving equipment.

Table 2.13 Comparison of methods for removing soil surface, contaminated in warm weather (29).

Condition of surface	Implement	Removal of radioactivity ¹	Effort required—		Effect on soil productivity ⁴
			For removal ²	For disposal ³	
Bluegrass sod	Soil cutter 12" wide	Good to fair	Poor	Fair	Good to fair.
Fescue-clover meadow	Vacuumized sweeper	Poor	Poor	Good	Good.
Fescue meadow	Rotating-broom sweeper	Fair	Fair	Good	Good.
Fescue-clover meadow	Motor grader	Fair	Poor	Poor	Good to fair.
Fescue-clover 12" high	Motor grader	Good to fair	Poor	Poor	Good to fair.
Soybean stubble	Motor grader	Fair	Poor	Poor	Good to fair.
Soybean stubble	Constant-draft scraper	Fair	Poor	Poor	Good to fair.
Wheat stubble	Vacuumized sweeper	Poor	Poor	Good	Good.
Corn stubble	Motor grader	Poor	Poor	Poor	Good to fair.
Flowed	Motor grader	Fair	Poor	Poor	Good to fair.
Flowed	Bulldozer	Good	Poor	Poor	Good to fair.
Flowed	Self-loading scraper, 1 cu. yd.	Fair to good	Poor	Fair	Good to fair.
Flowed	Pan-type scraper, 8 cu. yd.	Good	Poor	Fair	Fair.
Disked	Motor grader	Fair to poor	Poor	Poor	Good to fair.
Disked	Rotary scraper	Fair to good	Poor	Fair	Good to fair.
Disked	Elevating scraper	Fair	Poor	Fair	Good to fair.
Seedbed	Motor grader	Good to fair	Poor	Poor	Good to fair.
Seedbed	Bulldozer	Good to fair	Poor	Poor	Good to fair.
Seedbed	Self-loading scraper	Fair	Poor	Fair	Good to fair.
Seedbed	Pan-type scraper	Good	Poor	Fair	Fair.

¹ Rating of removal of radioactivity: Good—> 95 percent removal.

Fair—75-95 percent removal.

Poor—< 75 percent removal.

² Rating of removal effort: Good—> 5 acres per hour.

Fair—1 to 5 acres per hour.

Poor—< 1 acre per hour.

³ Rating of disposal effort: Good—additional loading and hauling effort minimal.

Fair—considerable effort in loading and hauling.

Poor—very great loading and hauling effort.

⁴ Rating of effect on soil productivity: Good—Increases or does not change productivity.

Fair—Reduces productivity < 20 percent.

Poor—Reduces productivity > 20 percent.

Studies on removal of surface soil have often shown some loss in soil productivity. Restoring the productivity of the treated area requires improvements in the physical structure and in the nutrient supply of the remaining soil.

In cold weather, the removal of surface contamination would usually be more difficult than in warm weather. If the soil surface was frozen, it could not be removed by scraping. Vacuum or sweeping machines might be useful unless the contaminant had been frozen into the surface.

Several methods removed 75% or even 95% of the contaminant that had been deposited on the ice or ground surface.

No effect on soil productivity would be expected from snow removal, and removal of thawing ground should have an effect comparable to that of removal of surface soil.

Table 2.14 Comparison of methods for removing soil surface, contaminated in cold weather (29).

Condition of surface	Implement	Removal of radioactivity ¹	Effort required—		Effect on soil productivity ⁴
			For removal ²	For disposal ³	
Loose snow 2 to 7" deep	Motor grader	Poor to good	Fair	Fair	Good.
Do.	Carryall scraper	Fair	Fair	Good	Good.
Do.	Bulldozer	Fair	Fair	Fair	Good.
Do.	Rotary snow blower	Fair	Poor	Fair	Good.
Loose snow 7 to 12" deep	Snow plow	Good	Good	Poor	Good.
Do.	Motor grader	Good	Fair	Poor	Good.
Do.	Carryall scraper	Fair	Fair	Fair	Good.
Do.	Rotary snow blower	Poor to good	Poor	Poor	Good.
Packed snow	Motor grader	Fair	Fair to poor	Fair	Good.
Do.	Rotary-broom sweeper	Fair	Fair to poor	Good	Good.
Do.	Vacuumized sweeper	Poor to fair	Poor	Good	Good.
Loose snow on packed snow.	Motor grader	Poor to fair	Poor	Fair	Good.
Frozen loose snow on packed snow.	Snow plow	Poor	Good	Fair	Good.
Frozen ground	Motor grader	Poor	Poor	Fair	Good.
Do.	Rotary-broom sweeper	Fair to good	Poor to fair	Good	Good.
Do.	Vacuumized sweeper	Poor to fair	Poor to fair	Good	Good.
Thawing ground	Motor grader	Good	Poor	Poor	Good to fair.
Do.	Carryall sweeper	Good	Poor	Fair	Good to fair.
Do.	Bulldozer	Good	Poor	Poor	Good to fair.
Do.	Rotary-broom sweeper	Poor	Fair	Good	Good.

¹ Rating of removal of radioactivity: Good—> 95 percent removal.
Fair—75 to 95 percent removal.
Poor—< 75 percent removal.

² Rating of removal effort: Good—> 5 acres per hour.
Fair—1 to 5 acres per hour.
Poor—< 1 acre per hour.

³ Rating of disposal effort: Good—additional loading and hauling effort minimal.
Fair—considerable effort in loading and hauling.
Poor—very great loading and hauling effort.

⁴ Rating of effect on soil productivity: Good—Increases or does not change productivity.
Fair—Reduces productivity < 20 percent.
Poor—Reduces productivity > 20 percent.

- Transuranides clean (TRUclean) process

A soil decontamination plant was developed in 1985 for the US Field Defense Nuclear Agency to remove plutonium contamination from coral and soil. The system was designed, fabricated and operated by AWC, Inc. (Las Vegas, Nevada) a Nuclear Services Corporation. Subsequent to its fabrication, testing and operation, the plant, known as the TRUclean Process, was declared a successful coral/soil decontamination process (38).

The pilot tests conducted on Johnston island during 1985 and 1986 by the TRUclean process opened ways for large volume reduction and activity removal. The tests demonstrated a somewhat greater than 90% volume reduction and activity removal from coral derived soil.

In 1986, the pilot plant was shipped from Johnston island to NTS and arrived at the Area 25 E-MAD facility.

A total of 132 drums of soil were shipped from the Rocky Flats Plant and delivered to the TRUclean test facility at the NTS, in 1987. The soil was typically a very dark fine grained clay with residual plant and animal matter, along with man made debris. Approximately 30% of the soil₃ particles were less than 150 μ in size. The bulk density was 1.076 g/cm³ and the specific gravity was 2.77 g/cm³.

Operational parameters were developed which enable them to remove greater than 90% of the incoming activity. They determined the plutonium content through the Am-241 measurement.

Final soil sample analyses confirmed that plutonium and americium were removed and a post-cleanup contamination level of 10 pCi/g of TRU was attained in all cases. It was expected, also, that this method will be able to lower radioactivity down to levels < 2.0 pCi/g (74 mBq/g) of Am-241 with a volume reduction of at least 90% (38).

- Clean-up and treatment (CAT) test: A vacuum method

The cleanup operations using the vacuum method of soil removal called Cleanup And Treatment test (CAT), was conducted by the Reynolds Electrical and Engineering Co. on Nevada Test Site and it has been presented in page 28.

2.3.3 PLOUGHING

2.3.3.1 Enewetak atoll

Decontamination efforts on large land masses in general involves soil removal. At Enewetak this poses an important problem: Little top sandy soil is available on these islands for the growing of crops; and, the islands are only slightly (1-3 m) above sea level. So, only ploughing or others methods of mixing soil bearing radioactive contamination was reasonable to develop in this atoll.

Janet island was selected for this experiment because it was the most important northern island of the atoll in terms of future residence,

agriculture or food gathering, depending on the final radiological status of the island after cleanup.

The site chosen for the ploughing experiment was relatively free of major debris and vegetation. It was located 300 m from one surface ground zero and 500 m from another. The total experimental area was 50 m x 100 m, divided into four parcels (1,2,3 and 4) 25 m x 50 m on the site. Rows 2 and 4 were to be ploughed; rows 1 and 3 were held as "controls" (5).

- Levels of application

In the experimental area of Janet island the surface concentration of ^{241}Am ranged from 14 to 32 pCi/g (518 to 1184 Bq/kg) of soil, with an average of 21 pCi/g (777 Bq/kg).

- Machinery

A 91-cm mold-board plough was pulled by a D-8K Caterpillar tractor. Unfortunately, the hydraulic ram on the plough failed and could not be repaired at Enewetak. Since the plough could not be raised or lowered hydraulically, a front-end loader was used to start the plough into the ground and lift it out.

Rows 2 and 4 were ploughed, lengthwise, and the plough was turned outside each plot, without taking it out of the ground. The operator experienced some initial difficulty in properly overlapping the furrows and in aligning the cuts to the plot. Occasionally, old cable turned up and was cleared away by lifting the plough out of the ground outside the experimental area.

- Efficiency

The plough had to be pulled at a fairly rapid rate (about 67 m/min) to turn the sandy soil over satisfactorily. After the areas were cleared of vegetation and debris, ploughing proceeded more satisfactorily. The plough was effective in ploughing to a depth of about 50 cm, even ripping through partially consolidated coral.

The americium-241 present in the soil was used as a tracer to determine the effectiveness of ploughing in mixing the soil from the surface to depth. Pre- and post-ploughing surface and profile measurements were made of soil types and Am-241 concentrations.

Ploughing was relatively effective in mixing ^{241}Am at the surface down to 50 cm, although "hot spots" were evident at various depths. Surface concentrations which averaged from 14 to 32 pCi/g (518 to 1184 Bq/kg) were reduced to 1.2 to 3.6 pCi/g (44.4 to 133.2 Bq/kg). The remaining surface fraction of Am-241 activity ranged from less than 5% to 25% with an average value of 14% of the initial surface contamination (33). Similarly, organic matter from upper levels of soil likely was mixed fairly well to lower depths, although the comparatively darker organic soil appeared here and there as thin layers in lighter colored coralline sand.

No decision was made about whether ploughing would be an acceptable technique for use in the cleanup program at Enewetak atoll. Accordingly, the contaminated surface soil was removed from the two unploughed plots of Janet.

2.3.3.2 Nevada Test Site

At the Nevada Test Site's Area 13 (Great Basin Desert) eleven 50 x 100 ft² (15.24 x 30.48 m²) test plots were established about 1000 ft (304.8 m) north of ground zero, for the decontamination studies in connection with the Pu fallout (Dick and Baker, 1967 (39)).

Ploughing, disking and scraping there were made combined with other treatments. Among the treatments were road oil, foam from fire fighting equipment and flooding. The scraping was with and without road oil, and with water. In one treatment, an FeCl₃ solution was applied, supposedly to induce cementing of Pu particles and soil particles in order to decrease the probability of resuspension (17).

- Levels of application

The higher mean activity of the areas where ploughing, scraping and disking treatments were carried out were 43.8, 20.7 and 8.3 Bq/m³ of soil, respectively.

- Machinery

The equipment used included a road grader, a simple-bottom farm plough, a fourgang disk harrow, an 850-gallon (3.2 m³) water-sprinkling truck, and two 400-gallon (1.5 m³) trucks.

- Efficiency

The radiactivity attenuation efficiencies of the different treatments for land areas are shown in table 2.15. Greatest attenuation of Pu-activity was achieved by ploughing (98% ?) and by oiling and scraping treatments (96%).

Table 2.15 Land area decontamination efficiencies
((39) referred in (17))

Method	Mean initial (dpm/m ²)	Mean final (dpm/m ²)	Effic. (%)
Plowing	2630	55	97.9
Oiling and scraping	1240	55	95.6
0.3-inch water leaching and scraping	205	15	92.7
0.3-inch water FeCl ₃ leaching	1405	118	91.6
Disking	500	54	89.2
1.0-inch water leaching	515	65	87.4
Scraping	79	11	86.0
Oiling (RC-0 road oil)	121	37	69.4
0.3-inch water leaching	8133	3660	55.0
0.3-inch water-Alconox leaching	380	309	18.7

Dick and Baker in 1967 (39) concluded, paradoxically, that ploughing and oiling were the most effective and cheapest of the various treatments used.

Water and fire-fighting foam treatments were effective for 1 hour in preventing resuspension and to a degree permanently because of pressure-induced mixing in the first few cm of soil. These can be looked upon as temporary countermeasures (17).

2.3.3.3 Palomares

- Levels of application

Watering, ploughing 30 cm deep, removing debris, raking, mixing and watering again of all lands showing contamination between 1200 kBq/m² and 1.2 kBq/m² were the countermeasures taken in order to dilute and diminish plutonium contamination from the surface of soil.

224 H₂ were ploughed to a depth of 30 cm, where contamination was less than 1200 kBq/m². Zones where contamination was greater than 1200 kBq/m² were ploughed after the 10 cm soil surface removal process.

- Efficiency

In order to evaluate the efficiency of this countermeasure, we consider the "dilution factor" which is obtained by dividing the final contaminated depth of soil and the initial depth.

If we consider the initial contaminated depth between 1 and 100 μ m (36), and the final one of 45 cm, the dilution factor is between 4.5×10^5 and 4.5×10^3 .

However, it was observed, at the time, that the concentrations onto the soil surface of the ploughed areas, were very superior to that derived from these dilution factors, due to the fact that an homogenization of soils has not been obtained during the last 20 years.

2.3.3.4 Chernobyl

Among the decontamination operations were a set of special agrotechnological measures which are recommended according to the soils and climatic conditions, the composition and type of radionuclides and other specific circumstances. The discharged radionuclides are practically insoluble in water and they are not very mobile when spreading through the biological chain soil-plants-animals-man.

To neutralize the short lived nuclides, it is enough to plough them into the soil, where they are bound, and await decay in a natural way. Applying any binding agent to the field before ploughing would help increase the efficiency of this operation.

The long lived isotopes could easily migrate from the soil to the organisms. Thus, it is not enough to plough them in, but, to bind them

chemically. That is why the plots contaminated by these isotopes should be treated with lime, zeolites or similar substances (40).

A range of agrotechnical and agrochemical measures designed to make agricultural products fit for consumption were implemented in contaminated regions of the Ukraine, Byelorussia and the Russian Soviet Federated Socialist Republic in 1986 and 1987. Deep ploughing was carried out and large quantities of inorganic fertilizer were applied to hundreds of thousands of hectares of contaminated land in these republics. Work was under way to ameliorate meadows and pasture land. Steps were being taken to reduce the transfer of radioactive substances from the soil to crops by applying lime, phosphoric and potassic fertilizer and certain sorbents (zeolite) to the soil. In the first year following their implementation, these measures have reduced the levels of radioactivity in agricultural products by a factor of 1.5-3. The full implementation of all the measures stipulated by the USSR State Commission for the Agricultural Industry would probably result in a substantial reduction in the exposure of the population from food (24).

2.3.4 OTHER FIELD RESEARCHES AND LABORATORY TEST

2.3.4.1 Deep ploughing of contaminated soil

The feasibility of several depths ploughing treatments is summarized in table 2.17. Ploughing, i.e. 36 inches (91 cm) deep, requires special machinery and the effects on strontium uptake may vary greatly with different soils and crops. Ploughing 7 inches (18 cm) or 12 inches (30 cm) has little effect on uptake of radioactive strontium. Only by using some material or technique to stop root growth into the contaminated soil could reduce effectively the radioactivity uptake (29).

2.3.4.2 Irrigation and leaching

Irrigation and leaching would not be very feasible treatment for reducing the uptake of radionuclides (tables 2.15 and 2.17), and the soil productivity would be lowered by leaching because essential nutrient elements would be removed with the contaminant.

In Nevada Test Site, also, during laboratory work with soil samples, plutonium which is associated primarily with silt size particles, was leached with 8 M nitric acid (1 h contact) and extracted with 0.1 M citric acid (30 minutes extraction) at room temperature (28°) (41). In these laboratory's selected leaching tests, the fractions leached in nitric and citric acid are shown in table 2.16. The low leachability in nitric acid and low solubility in citric acid of NTS samples would suggest a low uptake factor in plants grown in Nevada soils.

Table 2.16

Plutonium leachability of selected soils and sediments in
8 M nitric acid. One-hour contact at room temperature (41).

Activity Leached (Bq/g)	Total Activity in Sample (Bq/g)	Fraction Leached (%)
306	2666	11.5
31	210	14.7

Extraction of plutonium from selected soils
and sediments in 0.1 M citric acid.
Thirty minutes' extraction at room temperature (41).

Soil designation	Activity Leached (Bq/g)	Total Activity in Sample (Bq/g)	Fraction Leached (%)
Unsegregated	12	915	1.3
53 - 20 μ m	124	11084	1.1
20 - 5 μ m	37	3968	0.9
< 2 μ m	35	898	3.9

2.3.4.3 Minimum-tillage management

Where the soil surface contains most of the radioactive contamination, its uptake by crops could be lessened by growing deep-rooted crops under conditions of minimum-tillage. Estimated feasibility ratings for minimum-tillage management are given in table 2.17 (29).

2.3.4.4 Application of soil amendments

- Strontium uptake reduction

The effectiveness, effort, and productivity ratings of the application of lime, fertilizers and other soil amendments are summarized in table 2.17 for Sr-90. Unfortunately, none of these soil amendments are highly effective in reducing uptake of radioactive strontium (29).

- Cesium uptake reduction

Fertilization with potassium

Adequate fertilization with potassium, approximately 250 Kg K₂O/Ha per year, has proved to be one of the best measures to reduce Cs contamination of grassland forage. Cs-137 activity was 86 nCi per Kg forage (d.m.) on plots unfertilized with potassium and 27 nCi on plots fertilized well with potassium. The reasons are, without doubt, the well known antagonism between potassium and cesium on the one hand and the yield increase and dilution effect to be gained by fertilization with potassium on the other hand (42).

Table 2.17 Comparison of soil management methods for reducing Sr-90 uptake from contaminated soils (29).

Method	Reduction in Sr 90 uptake ¹	Effort required ²	Effect on soil productivity ³
Minimum tillage	Poor to fair	Good	Good to poor.
Plowing, 7" deep	Poor	Good	Good.
Plowing, 12" deep	Poor	Fair	Good.
Plowing, 30" deep	Fair to poor	Poor	Good to poor.
Plowing, 30" deep with root inhibition.	Good to fair	Poor	Good to poor.
Irrigation	Poor	Fair to good	Good.
Leaching	Poor	Fair	Poor.
Lime application, 2 to 10 tons/acre.	Poor to fair	Good	Good.
Nitrogen fertilizers, 100 # N/acre.	Poor	Good	Good.
Phosphate fertilizers, 100 # P/acre.	Poor	Good	Good.
Potassium fertilizers, 500 # K/acre.	Poor	Good	Good.
Organic compost, 5 to 20 tons/acre.	Poor	Fair	Good.
Clay minerals, 5 to 20 tons/acre.	Poor	Fair	Good to fair.
Ammonium or potassium phosphates, 2 to 5 tons/acre.	Fair	Fair	Fair to poor.

¹ Rating of reduction in Sr-90 uptake: Good—> 95 percent reduction.
Fair—75 to 95 percent reduction.
Poor—< 75 percent reduction.

² Rating of effort required: Good—Not significantly more than normal field practices.
Fair—Extra equipment, materials, or labor required.
Poor—Very great requirement of equipment, materials, or labor.

³ Rating of effect on soil productivity: Good—Increases or does not change productivity.
Fair—Reduces productivity < 20 percent.
Poor—Reduces productivity > 20 percent.

Fertilization with nitrogen and harvest time (growth stage)

The expectations to be able to dilute Cs-137 contamination of herbage by fertilization with Nitrogen were only partly fulfilled. Only if harvesting of the grass occurred relatively late (about in the "hay"-stage) clear differences in Cs-137 activity of the herbage were found: In fact, 26 nCi Cs-137 when unfertilized with Nitrogen and 15 nCi when fertilized with 60 Kg N/ha growth in form of calcium ammonium nitrate. No reduction in Cs-137 contamination was obtained by nitrogen fertilization, if the degree of contamination is relatively high and the grass is harvested at "pasture"-stage or earlier.

Cs-137 contamination can be diminished by delaying the time of harvest. In all levels of nitrogen fertilization, Cs-137 contamination of the grass was reduced by about 35 % by delaying the harvest time. The decontaminating effect of delaying the harvest time was much more pronounced when this measure was combined with ample nitrogen fertilization (42).

Liming

Only distinct overdressing with lime (1000 Kg/Ha CaO every second year) seemed to be able to reduce Cs-137 contamination of herbage (from 1.63 to 1.26 KBq/Kg d.m.) (42).

Plant species

There seems to exist material differences in Cs-137 activity in dependence on plant species (table 2.18) (42). The figures of this table may help to explain the results of farm scale measurements and could be advantageous for reseedling of grassland on areas heavier contaminated with cesium.

Table 2.18 Cs-137 activity in dependence on plant species (42)

Dactylis glomerata	23.6	Festuca rubra	46.4
Phleum pratense	17.3	Agrostis tenuis	16.6
Trisetum flavescens	11.9	Poa pratensis	10.7
Arrhenatherum elatius	8.3	Lolium perenne	7.5
Festuca pratensis	7.8		
Lolium multiflorum	7.6	Trifolium repens	19.9
		Medicago sativa	11.0
		Trifolium pratense	5.0

Experience from Chernobyl

As numerous studies have shown, among the most effective methods reducing Cs-137 uptake from soil by plants in the case of low fertility light textured acid soils are the application of mineral fertilizers (especially potassium fertilizers, considering the antagonism during the assimilation of Cs-137 and its chemical analogue potassium by root systems) and the liming of soils. Moreover, there are data indicating a decrease in the transfer of radionuclides (including Cs-137) to plants in light textured sward-podzolic soils when zeolite type clay minerals are added. In this case, the soil becomes enriched in finely dispersed fractions of clay minerals with a high exchange capacity, ensuring fixation of Cs-137 and its conversion into a state which is not easily assimilable by plants (43).

On the whole, addition of lime, mineral fertilizers and zeolites generally causes a reduction in the accumulation of Cs-137 in plants (table 2.19). The most effective reduction results from the addition to the soil of large amounts of potassium fertilizers (the greatest decrease was observed in the grain of oats in a soil of the sward-podzolic argillo-arenaceous silty sand type-by a factor of up to 3.5). On an average, in the case of grain, leaves and stems in all the plants studied, higher doses of potassium fertilizers lowered the transfer of this radionuclide to plants by factor of up to 1.5.

The addition of zeolites was effective only in one of the four soil types studied and only for particular crops: the greatest reduction in Cs-137 accumulation was detected in wheat (by a factor 2.5 in grain). In a number of variants the application of zeolite increased Cs-137 transfer to plants.

The effectiveness of the agricultural ameliorative measures depended on the biological characteristics of plants and the time of interaction of the amendments with soil. Thus, in the case of mustard (grain and straw) and maize (leaves and stems), the addition of lime, fertilizers and zeolite did not substantially lower Cs-137 transfer to plants.

Table 2.19 Coefficients of accumulation of ^{137}Cs in oats, wheat, mustard and maize (43)

Variants of experiment	Oats		Wheat		Mustard		Maize leaves and stems
	Grain	Straw	Grain	Straw	Grain	Straw	
1	2	3	4	5	6	7	8

Sward-podzolic sandy loam with a lighter sand fraction

Without fertilizer (control)	—	—	0.12	0.28	0.66	0.90	—
$\text{N}_{30}\text{P}_{30}\text{K}_{30}$ (background)	—	—	0.10	0.24	0.48	0.64	—
Background + $1\text{H}_2\text{CaCO}_3$	—	—	0.11	0.27	0.69	0.93	—
Background + zeolite 10 t/ha	—	—	0.11	0.27	0.42	0.72	—
Background + $1\text{H}_2\text{CaCO}_3$ + zeolite 10 t/ha	—	—	0.17	0.36	0.57	0.78	—

Sward-podzolic argillo-arenaceous silty sand

Without fertilizer (control)	0.042	0.11	0.030	0.08	0.18	0.24	0.07
$\text{N}_{30}\text{P}_{30}\text{K}_{30}$ (background)	0.027	0.07	0.024	0.05	0.14	0.19	0.06
Background + $1\text{H}_2\text{CaCO}_3$	0.030	0.08	0.021	0.05	0.15	0.20	0.05
Background + $2\text{H}_2\text{CaCO}_3$	0.024	0.06	0.018	0.04	0.14	0.18	0.10
Background + K_{120}	0.012	0.03	0.018	0.04	0.13	0.17	0.08
Background + P_{120}	0.027	0.07	0.015	0.05	0.16	0.22	0.07
Background + $\text{P}_{120}\text{K}_{120}$	0.015	0.04	0.009	0.02	0.17	0.23	0.07
Background + zeolite 5 t/ha	0.033	0.08	0.012	0.03	0.26	0.35	0.08
Background + zeolite 10 t/ha	0.039	0.10	0.012	0.03	0.20	0.25	0.09
Background + $1\text{H}_2\text{CaCO}_3$ + zeolite 10 t/ha	0.030	0.08	0.027	0.07	0.16	0.22	0.12
Background + $1\text{H}_2\text{CaCO}_3$ + K_{120} + P_{120} + zeolite 10 t/ha	0.027	0.08	0.021	0.05	0.14	0.20	0.07

Table 2.19 Continuation

Sward-podzolic light loam with a higher sand fraction

Without fertilizer (control)	—	—	0.15	0.36	0.78	1.10	—
$N_{70}P_{20}K_{30}$ (background)	—	—	0.14	0.33	0.84	1.14	—
Background + $1H_2CaCO_3$	—	—	0.14	0.33	0.57	0.81	—
Background + zeolite 10 t/ha	—	—	0.16	0.36	0.81	1.11	—
Background + $1H_2CaCO_3$ + zeolite 10 t/ha	—	—	0.17	0.39	0.54	0.72	—

Sward-podzolic argillo-arenaceous silty sand

Without fertilizer (control)	—	—	0.024	0.063	—	—	0.10
$N_{70}P_{20}K_{30}$ (background)	—	—	0.024	0.063	—	—	0.08
Background + $1H_2CaCO_3$	—	—	0.024	0.057	—	—	0.07
Background + K_{100}	—	—	0.015	0.042	—	—	—
Background + zeolite 10 t/ha	—	—	0.027	0.069	—	—	0.13
Background + $1H_2CaCO_3$ + zeolite 10 t/ha	—	—	0.027	0.069	—	—	0.10

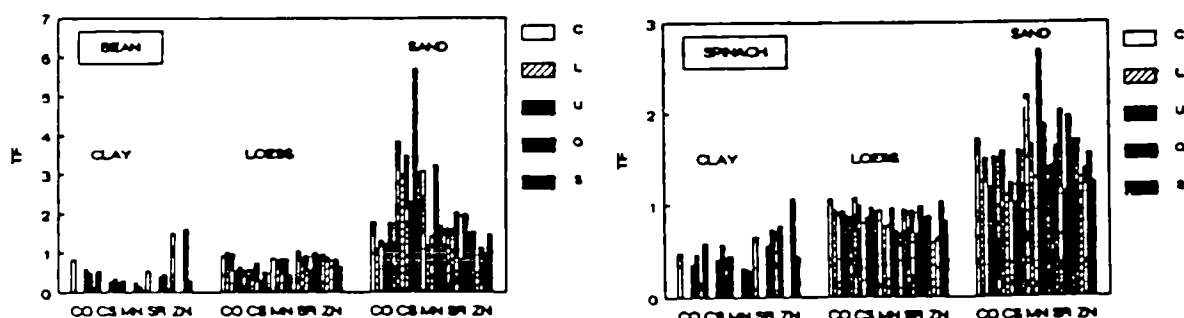
- Conclusions

Some general conclusions in relation to the use of soil amendments can be drawn:

- The TF's most affected by liming were those for Sr, Mn and Zn.
- Organic matter seemed to reduce transfer of Co, Cs and Sr and to slightly enhance that of Mn and Zn.
- No systematic changes were induced by varying the amounts of fertilizer added.
- Transfer factors are low on soils with a high clay content.
- Chemical amendments only affected translocation to foodstuffs.

The mean transfer factor for 5 elements and 2 crops grown on 3 soils as a function of 4 soil management practices (addition of lime (L), organic matter (O) and stable elements (S), omission of fertilization (U) and control (C)) is presented in fig. 2.2. The TF is expressed as a fraction of the global mean for the given crop (44).

Figure 2.2 Mean transfer factor for 5 elements and 2 crops grown on 3 soils as a function of 4 soil management practices (44).



The following measures, also, are promising for the grassland farmer to reduce Cs-137 contamination of the regrowth after a nuclear accident, like Chernobyl, according to the obtained results:

- (1) To adjust deficiencies of potassium in the soil.
- (2) To fertilize sufficiently with nitrogen - provided deficiencies of potassium are corrected - and to utilize the herbage relatively late (about at the "hay stage")
- (3) To reseed grassland with species which seem to have a low predisposition to contamination with Cs-137 like Lolium multiflorum, Lolium perenne and Trifolium pratense.

It may also be helpful to the farmer to know that Cs-137 contamination of the herbage is materially declining in the course of time (42).

2.3.4.5 Application of fixatives

The term "fixative" refers to any material used to bind radioactive particles to a surface (45). There are a variety of substances that could be used as fixatives, including a number of petroleum-based products such as road oil, emulsified asphalt, diesel oil, and MC-70.

Another group of materials includes those which are sold for the purpose of dust control and are sometimes called "dust palliatives", "dedustants", or "dust retardants". These include generic products such as calcium chloride, magnesium chloride and calcium lignosulfonate, and proprietary products such as Coherex and Compound SP.

In addition, there are other materials that could be used as fixatives even though that is not their primary function (i.e. a strippable coating); even plastic sheeting, paint or water could be used to prevent resuspension of radioactive particles. Decontamination foam, also, could be considered as a fixative, and it's described, separately, at the end of this section.

- Selection of the fixative entails several considerations, including:
- The physical characteristics of the fixative: Operation principle (forming membrane layers over the surface or binding the particles), toxicity (to plants or animals), cleanliness and removability, and durability.
 - The physical characteristics of the surface to be treated: Climate, surface nature, use of treated surface and environmental aspects.
 - The application technique.
 - The cost of the material and its transport to the accident site, and the cost of its application.
 - The effect of the fixative on ultimate cleanup procedures.

Table 2.20 presents basic fixative cost data on 1983, for the fixatives applied on ground surfaces (45):

- Road oils: Petroleum products designed as SCs (SC-70, SC-250 etc, higher numbers means greater viscosity). They are quite messy and have damaging environmental effects.
- MCs: These petroleum products are "cut-back asphalt"; that is asphalt diluted with kerosene distillate and therefore more viscous.

- Emulsified asphalt: Doesn't have to be heated and, because these asphalts are water based, can be easily and cleanly handled.
- Other petroleum products: Diesel, bunker oil, emulsifier dust. Their application could have serious environmental effects.
- Coherex: A proprietary product of the Witco Co. It is a liquid emulsion of petroleum resins and can be applied with a water spray truck.
- Compounds SP (-301 and -400): Products made by Johnson March, Inc. They are long-chain polymers which can be sprayed undiluted using an orchard sprayer or a water spreader truck.
- Calcium and magnesium chlorides: Hydrophilic products that form a liquid coating over the material on which they are placed, by attracting moisture from the air. In very arid areas these materials will quickly dried out and periodic application of water will be necessary.
- Lignins (polybinder and lignosite): By-products of wood pulp processing. They are not hydrophilic and can be used in arid areas. They are biodegradable (one year of durability) and non-toxic .

In the table 2.20, for each fixative, the dilution rates (using water as dilutant), the membrane or binder formation, the durability, the need of load and a rough indication of the prospects for removing the fixative (cleanup) are, also, presented (45).

Use of gravel and asphalt

In the drum storage field at Rocky Flats Plant₂ (with levels of Pu contamination in the soil ranged from 200 to 30000 kBq/m²) a gravel cover of approximately 15 cm deep was applied, and later the area was complementary stabilized by covering the gravel with a 7.5 cm deep asphalt pad (9).

Use of polyurethane foam

Polyurethane foam is an agent which would cover the contaminated surface and retains the contaminants until better conditions were available for its pick-up (46). The application of this countermeasure was studied at three sites of Rocky Flats Plant.

Three parameters were considered in choosing the test sites: Soil type, vegetation and surface condition of soil.

Test site A (a gravel fill, flat and without vegetation):

The normal rate of application was 1.4-1.9 m²/minute, in two passes, making a 5 cm thick layer.

Test site B (a soil-gravel surface):

Vegetation was sparing and required special attention.

It was covered at a rate of 0.9 m²/min.

Test site C (an undisturbed soil, the surface was inclined 35°):

The application of foam was accomplished using an inclined (= 45°) position gun.

- Machinery:

Two commercially available foaming units were used to deliver an uniform 5 cm layer of foam on soil. The first involved an air-driven pump transfer unit with a rate of surface coverage of 1.1 m² per minute ; this unit was not particularly reliable. The second spray unit was a pressurized foam transfer system with a surface cover rate of 0.9 to 1.8 m² per minute and an excellent reliability.

Table 2.20 Summary of fixative cost data, on 1983 (45)

Fixative Source	Price as sold	gal/unit	\$/gal	Dilution	\$/dil.gal	Applic. gal/yd ²	\$/yd ²	\$/m ²	Membrane (M) or Binder(B)	Durability (years)	Load?	Cleanup
Road Oil												
Wa.State Dept. of Trans.	\$160/ton	200 g/t	0.64	---	0.64	0.4	.256	0.31	B	20+	Yes	Very Difficult
Chevron	175-200 /ton	255-260 g/t	0.67-0.78	---	0.67-0.78	0.4	.268-.312	0.32-0.37	B	20+	Yes	
MCs:												
MC-100 Chevron	185-215 /ton	255-260	0.78	---	0.78	0.1-0.2 0.5=max	.078-.156	0.34	M		No	Very Difficult
MC-70 Shell	165/ton	252	0.65	---	0.65	0.1-0.5	.06-.325	0.07-0.39	110 ³ -135 ³ M		No	
MC-70 Representative						0.4		0.31	M		No	
Emul.Asphalt Chevron As.	135-150 /ton	240	0.56-0.62	var.1:0-5:1-10:1		0.20-0.25	.112-.155	0.134-0.185	M		No, unless thicker	Very Difficult
Shell	135-145 /ton	240 g/t	0.56-0.60				.224-.24	0.27-0.29	M			
Representative			0.60			0.25	.15	0.18	M			
Diesel Chevron	90-95/g	1	0.90-0.95	---	0.90-0.95	0.20-0.25	.18-.24		B			Difficult
Bunker Oil Chevron	26-30 /bbl	42 g/bbl	0.62-0.71	---	0.62-0.71	0.20-0.25	.124-.178		B			Difficult
Emulsifier Dust Oil - Chev.	150/t	240 g/t	0.625	---	0.625	0.2	.125	0.149	B			

Table 2.20 Summary of fixative cost data, on 1983 (Continuation)

Fixative Source	Price as sold	gal/unit	\$/gal	Dilution	\$/dil.gal	Applic. gal/yd ²	\$/yd ²	\$/m ²	Membrane (M) or Binder(B)	Durability (years)	Load?	Cleanup
Coherex Witco	.95/g	1	0.95	1:5 w	0.158	0.75	.118	0.142	B Hydrophilic	.5, 1.0*	Yes	Can be removed w/ petroleum solvent, will stain
Compound SP-301 Johnson March	2.15	1	2.15	---	2.15	0.09 g/100 ft ²	.1935	0.23	M	1	No	Difficult, binds like paint, clear
Compound SP-400 Johnson March	3.95	1	3.95	---	3.95	0.09	.3555	0.425	M	3	No	
Calc.Chl. (Pelladow) Van Waters & Rogers	.1317 /lb	---		5 lbs/gal 38% sol	0.6585	0.27	.1778	0.212	B Hydrophilic	.5 (?) w/load	Yes	Can be washed off
Liquidow Dow	100% 275/t	182	1.51	38%	0.573	0.6	.344	0.412	B	.125	Yes	
Mag.Chl. Burris Oil	.50/g	1	0.50	1:4 20%	0.10	0.5	.05	0.060	B Hydrophilic	.25	Yes	
Polybinder Burris Oil	.80/g	1	0.80	1:1 50%	0.40	0.5	.20	0.239	B	.25	Yes	
Polychem	.75	1	0.75	1:1 50%	0.375	0.5	.1875	0.224	B	1.0	Yes	
Lignosite Geo.Pac.	38/ton	190	0.20	1:1 50%	0.10	0.5	.05	0.06	B	1.0	Yes	Can be washed off w/ water until it dries, then use oil solvent
Yakima Co.	28.50 /ton	194	0.1469	3:1 w .75%	0.1102	0.37	.041	0.05	B	1.0	Yes	

* Coherex - First application = 6 months. Thereafter, 1 year. This is for surfaces with load.

Lifting of the hardened foam from the soil was done easily by either manual or mechanical means. Pick-up of sprayed soil was good because soil particles become encapsulated in the foam.

- Efficiency:

In a soil-gravel surface with contamination upward to 0.6 mCi/m^2 (22.2 Bq/m^2) about 85% of the activity was removed by a single application of foam 5 cm deep over the soil.

- Cost:

Application of a 5 cm thick coating foam, which adequately stabilizes the soil and is optimum for handling during disposal, requires 1.7 kg/m^2 for relatively flat surface. Cost material was about $2.4 \text{ \$/m}^2$ (on 1973).

- Practicability:

The foam is comparable to an asphalt cover but no surface preparation is required prior to foaming. Expense and complexity of foam-laying equipment is considerably less than for other kinds of durable coverings.

The durability of the foam to weathering was excellent. Minor deterioration of the foam was noticeable after 10 months. Expected duration of unprotected foam is approximately 18 to 24 months. No adverse effects, other than smothering, were observed on vegetation.

Plants which were covered totally by the foam during spraying were killed. Plants which were only partially covered survived, and even flourished, showing increased growth rates relatively to surrounding plants.

2.3.4.6 Screening, attrition scrubbing and cation flotation

Several soil conditions exist at Rocky Flats that are advantageous to decontamination processes. First, the soil is very rocky, and the disperse contamination exists on the surface of the minerals. The surface-contaminated soil contains only 20% clay and organic material and the particulate plutonium oxide is small in particle size (47,48,49,50).

Four processes were investigated to decontaminate Rocky Flats Plant soil to $< 30 \text{ Bq/g}$; the soil contained up to 45000 Bq/g soil, as Pu-239. The processes were:

- 1) Wet screening at high pH.
- 2) Attrition scrubbing at high pH.
- 3) Attrition scrubbing at low pH.
- 4) Cation flotation

1) Simple wet screening decontaminates 60 to 70 wt% of the soil. By adjusting the pH to 11, 76 wt% of the soil was decontaminated by wet screening to less than 30 Bq/g . The material was returned to the environment. The remaining material (30 to 40 wt%) either would have to be subject to further decontamination or packaged and shipped away for storage. The decontaminated material represents the fraction greater than 0.42 mm in size.

2) Calgon^(R) solutions at high pH, also, effectively decontaminated RFP soil. The soil is scrubbed in a rotary-type attrition scrubber (jar mill)

four times with the fines being decanted each time. Approximately 80 wt% of the soil is decontaminated to 30 Bq/g. This represents removal of 99.9% of the activity from the decontaminated portion.

3) Attrition scrubbing at low pH is also effective. An aqueous solution of 2% HNO_3 , 0.2% HF, 2% pine oil and 5% Calgon used in a rotary-type scrubber decontaminates 84 wt% of the soil to less than 5 Bq/g. Originally the soil contamination was 45000 Bq/g.

4) The fourth decontamination process takes advantage of the anionic surface of the clay particles. A cationic flotation agent such as an amine can be used to float the clay material in a conventional flotation process.

Efficiency

Some test were made to compare decontamination results on different soils. Results indicate that high pH solutions only remove contamination by hydraulic grading and attrition; the dilute acid (i.e. 2% HNO_3) removes soluble sorbed species and particulates by dissolution and attrition; and the 2N-HCl solution serves to remove more insoluble species by dissolution and attrition, and also leaches contamination that has diffused into the soil constituents (including rock).

Practicability

The attrition scrubbing process at high pH was the most feasible process to scale up. It was considered that several areas need more research before final design criteria could be issued.

2.3.5 WINDSCALE: MILK CONSUMPTION RESTRICTION

Radiochemical analysis of milk samples available, in October 1957, indicated contamination of milk by iodine-131, and a decision was taken to prevent the human consumption of milk. This restriction was extended to all areas where the activity of radioiodine in milk exceeded 0.1 $\mu\text{Ci/l}$ (3700 Bq/l). The total area was 200 square miles (518 km^2), covering a rectangular strip of coastline about 16 km wide, from 10 km north of Windscale southwards to the Barrow peninsula (7).

On the advice of the British Medical Research Council the conditions for derestricting an area involved demonstrating that:

- all milk samples had an activity less than 0.1 $\mu\text{Ci/l}$ (3700 Bq/l), and
- the decrease of the activity in successive samples showed a half-life of not more than 8 days.

This was achieved within 25 days for the most of the region, although in the highest contaminated area, close to Windscale, the period of the milk consumption restriction was extended to 44 days.

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3. AGRICULTURAL PRACTICES

3.1 DISTRIBUTION OF DIRECT CONTAMINATION FROM AN AEROSOL DEPOSITION ON VEGETATION AND SOIL

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3.1.1 INTRODUCTION

In compliance with the terms of CEC contract B 16-0265 F, this document summarizes the work undertaken as part of the "post-Chernobyl" program covering fallout distribution between the soil and cultivated crop cover. The experimental work comprised a phase in which cultivated plots were labeled with a micron-scale aerosol, followed by analysis of the tracer deposited on the plants and soil.

The experimental techniques are first briefly described before presenting the results, followed by a discussion and the principal conclusions and limitations applicable to the experimental data.

3.1.2 MATERIALS AND METHODS

Aerosol

The granularity had to meet several criteria, including similarity with the Chernobyl aerosol (H.BONKA, 1987), measurability, nontoxicity and rapid deactivation (to allow reuse of the test site within a short time).

The most suitable product was found to be a 25 g.l^{-1} aqueous solution of uranine dispersed by means of a commercial ultrasonic generator and measurable with adequate sensitivity ($1 \times 10^{-7} \text{ g.l}^{-1}$) by fluorimetry. The resulting aerosol had a mean mass diameter of $1.5 \text{ }\mu\text{m}$ with a standard deviation of 2.2, corresponding to a suitably monodispersed aerosol.

Labeling

The test carried out under this program involved only the dry deposit.

The method initially implemented consisted in moving a mobile "tent" containing the aerosol generator along a regular pattern over the vegetation. This method avoided wind interference, but resulted in an unrealistic vertical concentration profile.

Beginning in early 1988, the labeling was performed in the open with the aerosol generators mounted on a dolly upwind from the crops. Testing showed that the vertical concentrations obtained in this way were homogeneous to a soil depth of three meters, provided the generator was placed at least thirty meters upwind from the crops.

This technique allowed us to step up the test rate considerably: 19 test were conducted in 1988 and 27 in 1989, compared with a total of only 10 for the first two years. Combined with an elaborate sampling strategy, the method provided extremely satisfactory statistical results, reducing the confidence intervals by nearly a factor of 5.

Samples

The deposition on the plants was assessed using whole plants.

Soil deposits were evaluated using fallout gauges (stainless steel disks 20 cm in diameter) placed directly on the ground. The advantage of this method was to allow nearly 100% uranine measurement efficiency; with soil sample measurements the results fluctuated unpredictably between 40 and 60%, and could not be used. Prior testing showed that the gauge measurements were representative of the deposition on the cultivated soil.

Measurement procedures

The samples were rinsed in a known quantity of buffer solution (pH 9.4), filtered to 0.45 μ m and measured by fluorescence (513 nm). The fluorimeter was calibrated to allow the deposit quantities to be calculated.

Plant specimens

We determined the crops representing the largest cultivated surface areas in European countries. The test were conducted at Cadarache in Mediterranean climatic conditions, precluding the selection of certain species requiring cool temperatures or high humidity.

The following plants were tested: grains (wheat and corn), oleaginous plants (rapeseed and sunflowers), textile plants (flax), market garden plants (tomatoes and lettuce), industrial vegetables (peas, beans, potatoes, cabagge and Brussels sprouts) and forage crops (beets, alfalfa and meadow grass).

3.1.3 RESULTS

Table 3.1.1 indicates all the test results, expressed in terms of productivity (dry weight per square meter of crop land) and of the plant growth stage, identified by letter:

- A: seedlings
- B: growth or cell elongation
- C: flowering
- D: commercial maturity or fructification.

Sufficiently homogeneous results were available for some species to plot the plant catchment coefficient versus the productivity. (Figures 3.1.1 to 3.1.4)

3.1.4 DISCUSSION-CONCLUSIONS

The test data show significant variations in the catchment coefficient depending on the plant species, the growth stage and the crop

density. When estimating damage or evaluating countermeasures, a single graphic or mathematical relation is thus insufficient. We therefore propose a graph for each species (or group of related species).

This work must of course be continued to compile complete data on all widely cultivates species. For example, vineyards and orchards and forest cover have not been considered to date. In addition, now that the technique has been proven, it will have to be adapted to an investigation of moist deposits.

This study therefore qualified an experimental technique that will continue to be used in the future. It also demonstrated its applicability in improving strategies to protect environmental and economic interests in the event of a major accident involving a nuclear facility.

3.2 SOIL TREATMENT

3.2.1 CHEMICAL

STUDY OF THE POSSIBILITY OF ATTENUATING SOIL-PLANT TRANSFER AFTER AND ACCIDENT, BY APPLICATION OF MANURE TO THE SOIL AND BY FOLIAR SPRAYING

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3.2.1.1 Introduction

The respective effects of calcium and potassium on the transfer of cesium and strontium have been mentioned in the literature (1). A universal method describing how to use these chemicals in order to reduce the transfer factor of radionuclides, do not exist to our knowledge. Without claiming to attain the result, this study aims at defining conditions for choice of known chemicals and at determining the possible influence of unrecognized chemicals. The first part of this study is based on a correlation analysis of the transfer factor and soil parameters of different soils types fertilised in the course of greenhouse experiments. The second part is devoted to specific studies of each element N, P, K in various conditions. Therefore, the study of an other method for attenuating the soil-plant transfer will be presented such as the foliar application of fertilisers.

3.2.1.2 Material and methods

Cultures for correlation analysis.

Different soil types, pH 6-8, OM 1-10%, coarse sand 4-80%, the most frequently found around French nuclear sites, are contaminated by a mixture of cesium 134 as carbonate (specific radioactivity: 0,1MBq per microgram) and strontium 85 as carbonate (0,3 KBq per microgram) in an aqueous solution. The contaminating solution is poured onto a soil layer of about 2 cm. The soil contamination is homogenized in order to reach the activity of 0,3 Bq/g of soil, for each radionuclide. Culture post contain 10 Kg of soil.

Plants cultivated in a climate-greenhouse, belong to a list of species representing the cultures most frequently found around French nuclear sites (pasture, wheat, colza, potato, sunflower, carrot, bean). They are watered with a nutritive solution (COIC-LESAINT, 14,4 milliequivalent of nitrogen per liter).

The radioactivity of the edible part of the plants is measured by a pure Ge detector (10% of relative efficiency, 1,7 Kev of resolution) and a TN 7200 analyser of 2048 channels adjusted to an energy range of 600 kev which can detect 0,5 Bq per sample.

Soil analyses are made according to standardised methods (2).

The measurement of the cesium fraction retained by the soil is carried out by counting water extracted from the soil through a 0,45

micrometer filter membrane. Operating conditions necessitate the absorption of the soil solution when it fills 70% of dry soil volume. Sodium polycrylamide granules which can absorb 40 times their volume of water must be placed over the membrane.

Cultures enriched with manure, N, P, K and lime.

Manure: The soil samples previously studied are enriched with potassium sulfate in a first experiment, and with ammonium phosphate in a second experiment. Each soil type is fertilised at the rate of 4 concentrations in a range established from the natural level to 2, 4 and 6 times greater. The contamination is carried as similarly described before except for the culture of beans contaminated by cesium only and realised on the previous soil types and on compost (Triohum P, KLASMAN) in order to study a soil rich in organic matter.

Liming: Three soil types (sand: 4, 50, 80%, limestone: 45, 0, 0%), contaminated with cesium 134 and strontium 85 have been enriched with calcium carbonate at the rate of 0, 10, 25 g/kg. This experiment has been reproduced on heath mould (pH = 3,75) enriched either with calcium hydroxyde (0, 1, 4, 8 g/kg) as calcium carbonate (0, 1, 10, 25 g/kg). Each pot (20*15*10 cm³) containing 2 kg of soil is scattered with grass (ray-grass 50%, fescue 40%, meadow-grass, 10%) or French beans. Plants are grown in greenhouse under the same conditions as in the previous experiment except for the fact that the pots are watered with tap water. After three weeks the grass and bean leaves are harvested, washed, dried, and the radioactivity measured.

Correlations analysis.

The correlation of the transfer factor and the chemical and physical properties of the soil are analysed with the help of a step-by-step multiple linear regression program. The transfer factor values relative to each plant species are divided by their mean so that the regression calculation carried out with all plants blended, only integrates variations due to soil type or agronomic processes.

Foliar spraying of chemicals.

In a first experiment, bean cultivations on compost contaminated by cesium 134 under the conditions described previously are sprayed with an airbrush by solutions of different salts in 4 concentrations: 0, 0.5, 1, 1.5, g/l of a volume of 2 ml for 10 plants at the stage of first trifoliate leaves. The salts are calcium and magnesium sulfate, monoammonic and trisodic phosphate, cesium iodide and hydroxyde and potassium nitrate and acetate. Anions are chosen so as to obtain two different levels of solubility of the salt. In a second experiment, the concentration range is raised to: 0, 5, 10, 20 g/l (higher concentrations were phytotoxic). The solution is sprayed at the rate of 20 ml per plant. Potassium phosphate, nitrate, are sprayed in the same conditions, with the addition of an abrasive (5 g/l of silicium carbure), of a moisturizing solution (from alkylphenol oxyethyl 0,5 g/l), of ionophore (valinomycin, nonactin, in the vaseline oil dropped on to the leaves).

These additions are to facilitate the penetration of potassium in an aqueous solution through the double barrier created by the waxy cuticle, and the membranes of foliar cells constituted by hydrophobic compounds. Three

other plants were tested with potassium phosphate of 20 g/l, and different additions: colza, sunflower, potato. After the measurement of cesium 134 radioactivity, a sufficient number of bean leaf and pod samples were incinerated at 600°C to determine by atomic absorption, the proportion of potassium contained in the ashes, in solution in 0,1N nitric acid.

3.2.1.3 Results

Transfer factor correlations.

The multiple linear regression applies better to the Napierian logarithm than to its value. Therefore the correlations presented here concern the Napieiran transfer factor logarithm.

Exchangeable potassium and exchangeable calcium negatively correlate fairly well with the cesium and strontium transfer factor. (Table 3.2.1).

Table 3.2.1 Transfer factor and soil element analysis
correlations higher than 0.4

Correlation coefficient and limits for 0,95 security coefficient

	Cesium 134	Strotium-85
Exch. potassium	-0.44 +-0.20	-0.48 +-0.20
Exch. calcium	-0.65 +-0.15	-0.64 +-0.15
Coarse sand	0.68 +-0.14	
CEC	-0.60 +-0.16	
pH	-0.62 +-0.15	-0.60 +-0.16
Kd	-0.69 +-0.14	
Hydrosoluble Mn	0.66 +-0.14	
Saturation moisture		-0.63 +-0.15

This negative correlation is particularly confirmed for grass. (fig.3.2.1). Hydrosoluble potassium and hydrosoluble calcium are less well correlated. Other parameters like coarse sand content, pH, cationic exchange capacity and the sorption coefficient Kd, reciprocally correlated explain the transfer factor variability.

Fertiliser influence on the transfer factor.

Potassium: For most of the soils studied, the addition of 1g of potassium per kg. of soil leads to a reduction of the cesium transfer factor to the beans of an order of magnitude (fig.3.2.2). However, potassium addition in soil with low cationic exchange capacity leads firstly to a decrease and then to an increase of the transfer factor. In soils with high cationic exchange capacity, the transfer factor can be divided by 100. The quantity of potassium added to soil that leads to a reduction of the cesium transfer produces factor of an order of magnitude, also produces a reduction of that of strontium by a factor of 4.

Ammonium phosphate: The addition of 400 mg/kg of soil of biammonic phosphate leads to an increase in the transfer factor of cesium of an order of magnitude (fig. 3.2.3). It has no influence on the transfer of strontium.

Calcium carbonate and hydroxyde: Liming with 100 t/ha (25 g/kg) of calcium carbonate or 32 t/ha of calcium hydroxyde has no significant influence on the cesium and strontium transfer factors. (Table 3.2.2).

Table 3.2.2 Transfer factor of strontium 85 and liming intensity

Added Ca carb. g/kg	0	1	10	25
heath mould Ft (fresh pod/dry soil)	0.15 0.13	0.15 0.09	0.11 0.06	0.07 0.09
non calcareous soil Ft (fresh leaf/dry soil)	2.75	3.00	1.80	1.80
Calcareous soil Ft (fresh leaf/dry soil)	0.64	0.66	0.76	0.71

Foliar spraying of chemicals

Among the salts observed, potassium nitrate only is able to reduce the radioactivity of beans pods by 2. Experiments attempted in order to improve this result were vain. The potassium contents are not significantly higher than the contents of non sprayed leaves. The same was true for pods.

3.2.1.4 Discussion

The addition of K to the different soils with growing beans justifies the assumption concerning the reducing influence of potassium on the transfer factor of cesium already observed by correlation analysis. A similar study made by Sandalls, Eggleton and Guadern (3) indicates a positive correlation of the transfer factor and exchangeable potassium in the soil. These results must be compared to those obtained with the soils with a low cationic exchange capacity (< 120 me/kg)(fig. 3.2.2), and the soil rich in organic matter which was the case of the soils studied by these authors. In these conditions, the transfer factor increases from 1g/kg of added potassium to the soil. The cationic exchange capacity expresses the number of sites available for cation fixation. In the case where the ratio number of available sites/number of cations is inferior to 1, there probably, a cation exchange occurs which is not perceptible when the ratio sites/cations is superior to 1. The potassium thus desorbes the cesium increasing its availability for the plant. The same phenomenon arises with ammonium when ammonium phosphate is added inducing an increase of the availability of the cesium exchanged by ammonium.

The decrease of the transfer factor of cesium seems due as indicate Sandalls, Eggleton and Gaudern, to a reduction of the molar fraction of cesium in the alkali metal pool when potassium is added to the soil.

The correlation of exchangeable calcium to the transfer factor is not confirmed by specific experiments. Exchangeable calcium is correlated to coarse sand level, pH, and cationic exchange capacity, parameters which explain the variability of the transfer factor. This case shows that a correlation of two variables does not mean the influence of one variable on the other. The expected phenomenon of reduction of the molar fraction of strontium in the alkaline earth pool following the addition of calcium to the solution does not occur.

The spraying of potassium onto the leaves of the 4 plants under investigation did not replace massive supplies of potassium to the soil which in fact remain the most efficient method. The difficulty lies in supplying the necessary quantity of potassium that must be added in order to significantly exceed the quantity absorbed by the roots, without causing phytotoxicity. Those experiments did not lead to an increase of the natural potassium content of bean pods. It is therefore not possible to check if such an increase could influence the transfer of cesium.

3.2.1.5 Conclusion

These experiments show that potassium is the only element that can be used to reduce the transfer factor of cesium and to a lesser degree strontium. In some soils the potassium influence can involve a decrease of the transfer factor from 1 to 100. This countermeasure in case of an accident, could avoid banning cultivation in a contaminated field if the soil is rich in potassium and has high cationic exchange capacity. If the first requirement is not satisfied, a large quantity of potassium manure can be added. Ammonium fertilisers must be proscribed in case of an accident.

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Table 3.1.1

Plant	Stage	Yield (g·m ⁻²)		Dry/Fresh	Plant Catchment Fraction	
		Dry	Fresh		Deposit %	±
Wheat	A	130	350	0.37	53.0	16.4
	B	684	2780	0.25	66.0	34.9
	B/C	323	1747	0.19	74.6	2.9
	C	837	1690	0.50	67.0	3.6
	C	304	460	0.66	40.0	16.7
	C	323	1747	0.19	74.6	2.9
	C	839	1690	0.50	67.0	3.6
	C	1062	4000	0.27	58.3	3.8
	C	963	5157	0.19	76.0	1.7
	D	1017	1284	0.79	78.3	4.2
	D	1228	2738	0.45	66.0	32.0
	D	1017	1284	0.79	78.3	4.2
Meadow grass	B	492	1171	0.42	73.0	40.5
	D	354	715	0.50	87.5	3.9
	D	166	411	0.40	38.5	6.4
Corn	A	97	649	0.15	60.6	6.1
	A	42	283	0.15	40.9	6.5
	B	47	319	0.15	10.5	6.1
	B	212	1610	0.13	47.2	4.6
	B/C	432	1790	0.24	76.0	2.8
	C	637	4240	0.15	90.5	39.7
	D	1666	4200	0.40	83.8	5.8
Sunflowers	A	15.3	89	0.17	21.4	2.5
	A	40	240	0.17	37.0	7.5
	A	15	89	0.17	21.4	2.5
	A/B	40	240	0.17	37.0	7.5
	B	73	529	0.14	63.3	7.0
	B	58	426	0.14	40.5	18.6
	B	109	1368	0.08	26.2	9.0
	D	598	789	0.76	39.0	13.8
	D	350	1530	0.23	62.0	10.0
Tomatoes	D	605	4500	0.13	82.3	37.7
Peas	A	9	67	0.14	26.7	1.9
	B	35	129	0.27	38.5	3.8
	B	10	62	0.16	16.8	1.9
Beans	B/C	47	245	0.19	31.0	4.0
	D	283	1453	0.19	60.0	4.2
	D	258	1600	0.16	38.9	6.0

Table 3.1.1 (cont.)

Plant	Stage	Yield (g·m ⁻²)		Dry/Fresh	Plant Catchment Fraction	
		Dry	Fresh		Deposit %	±
Rape	B	750	5080	0.15	82.5	2.0
	D	1240	5210	0.24	80.2	6.3
Cabbage	A	5	29	0.18	5.6	1.2
	A/B	39	235	0.16	28.9	3.3
Brussels sprouts	B	44	337	0.13	29.6	4.6
	D	-	2110	-	48.7	4.4
Potatoes	A	10	51	0.20	14.9	1.3
	B	39	241	0.16	43.2	3.8
	B/C	131	938	0.14	59.8	6.8
	C	205	1170	0.18	70.8	7.1
	C	220	1525	0.14	70.2	8.9
	D	-	216	-	26.3	5.4
Lettuce	B	38	400	0.10	19.8	2.2
	B	25	315	0.08	19.2	1.5
Beets	B	170	1975	0.09	53.4	4.9
	C	156	1020	0.15	54.6	4.3
Alfalfa	C	-	501	-	64.2	3.6
	C	262	1509	0.17	89.6	1.6
	C	344	1504	0.23	89.7	1.4
	D	359	2323	0.15	95.1	1.4
Flax	C	71.5	296	0.24	71.8	6.3
	C	-	71	-	70.8	3.2
	C/D	242	726	0.33	67.8	3.3
	D	301	329	0.91	56.4	3.9

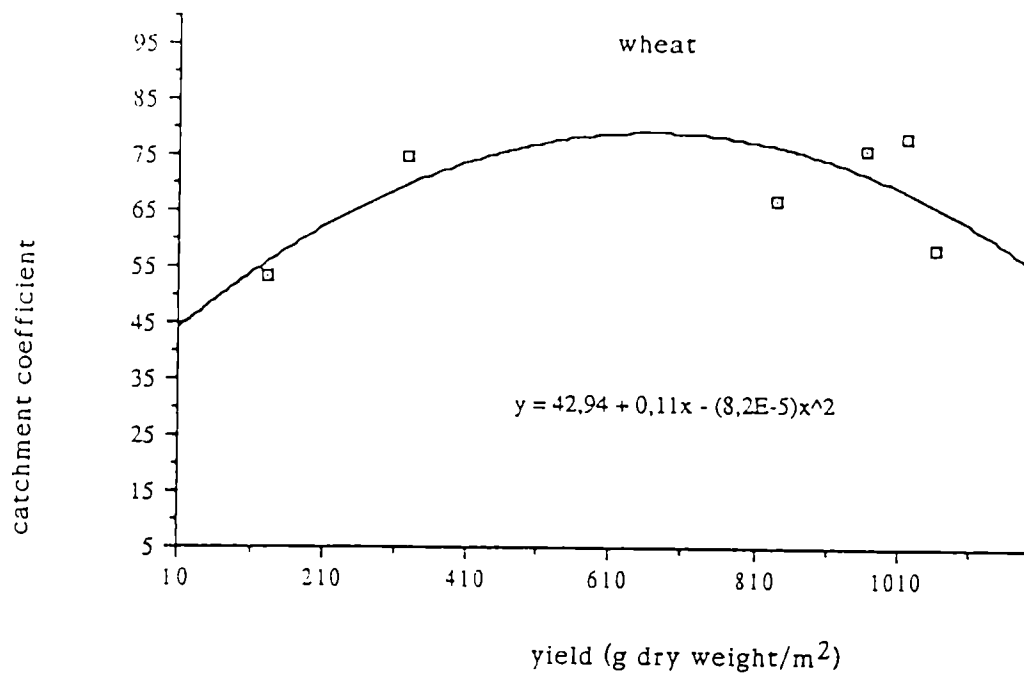


Figure 3.1.1

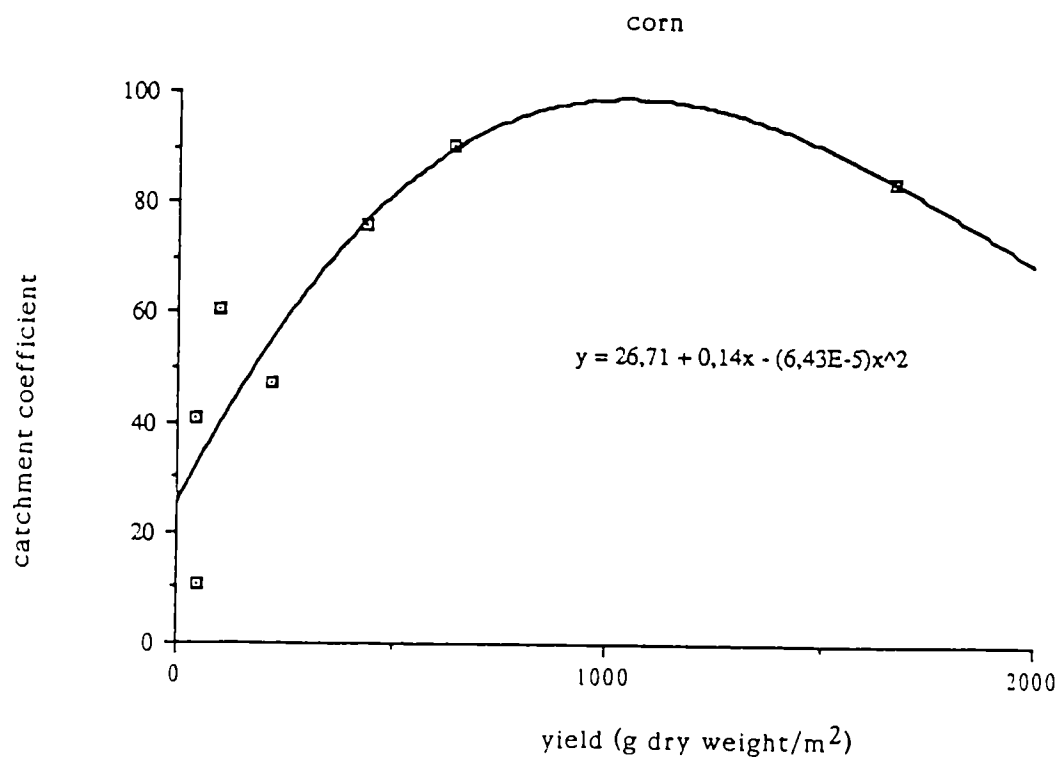


Figure 3.1.2

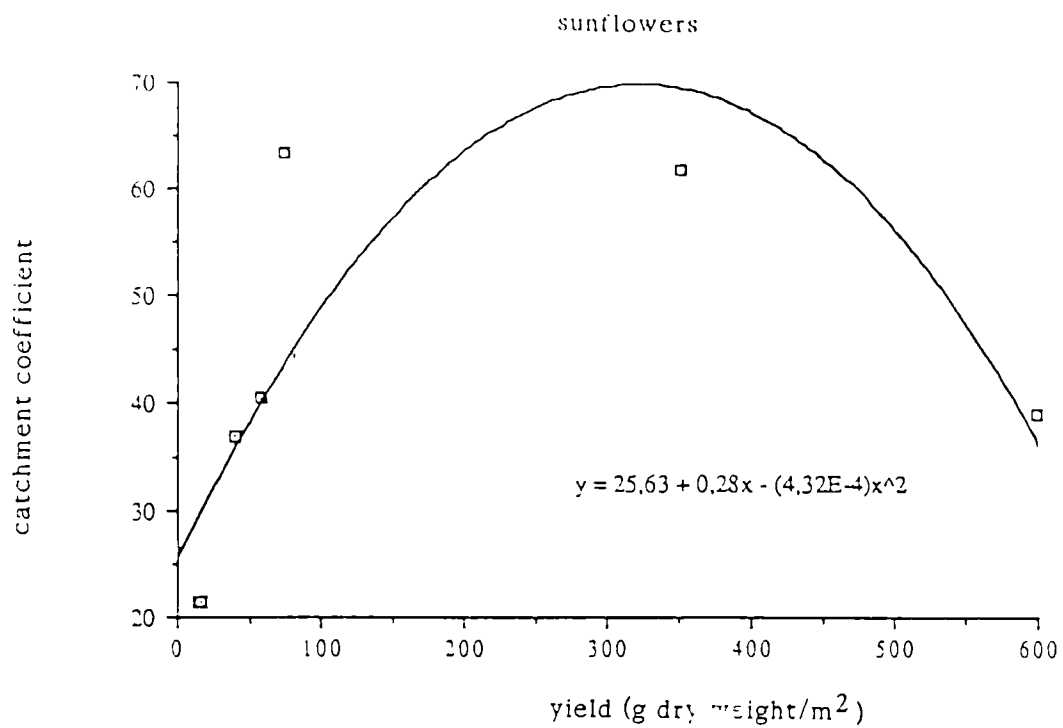


Figure 3.1.3

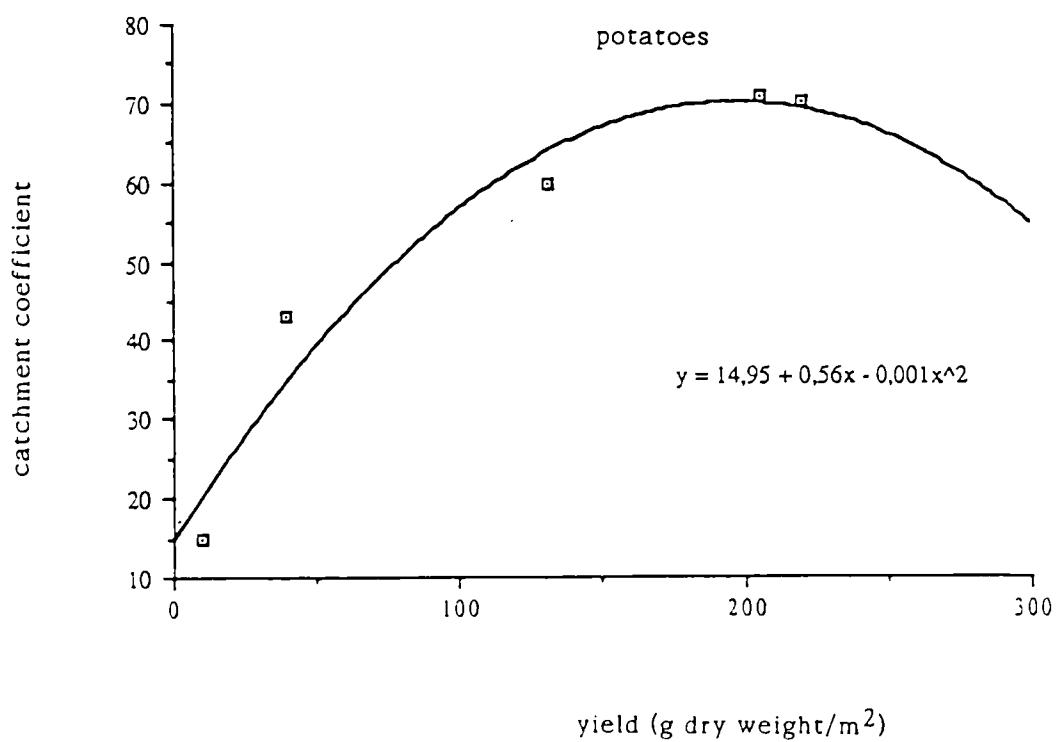


Figure 3.1.4

3.3 CROPS SELECTION

ROOT ABSORPTION FACTORS FOR VARIETIES OF CROP SPECIES

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3.3.1 INTRODUCTION

The contamination of vegetation with nuclear debris in connection with accidents in nuclear facilities takes place in two ways:

- 1) Direct contamination, i.e. adsorption on the aerial parts of the vegetation often followed by an absorption, and
- 2) Indirect contamination, i.e. absorption through the root system of radionuclides that have entered the soil.

While all radionuclides in nuclear debris play a role in direct contamination, it is among the long-lived nuclides, that absorption via the roots can be substantial.

Part of the Danish study was through a literature study to get an overview of the present days knowledge on root uptake of radiocesium, radiostrontium and plutonium by crops and from this study to identify possible countermeasures in order to reduce the long term effects of radioactive contamination of arable land. The results of this literature survey are reported by Beate Nielsen and Morten Strandberg (1).

The literature study was supplemented by experimental studies and field studies. The main results of these studies are reported in the next chapters.

3.3.2 EXPERIMENTAL STUDY CONCERNING INDIRECT CONTAMINATION

The aim of this part of the project was to identify crops with a relatively low or high root uptake of radiocesium.

3.3.2.1 Experimental methods

The experiment was carried out at Riso National Laboratory. The experimental methods used are described in details previously (2).

Four different varieties of barley and three different varieties of rye grass were seeded in 1988. Each crop was grown in two types of soil, an organic clay loam and an organic soil, both contaminated with radiocesium. Four replicates of each variety were made systematically throughout the experiment. In 1989 this part of the experiment was continued. Another set of pots containing clay-loam contaminated with radiocesium was seeded in 1989 containing two additional varieties of barley in order to confirm the results obtained in 1988.

3.3.2.2 Results

3.3.2.2.1 Barley.

The results for barley grain are shown in Table 3.3.1 The results are normalized to the mean of the concentration ratios for each category, A-C1, and for each year.

The root uptake for four different varieties in two types of soil contaminated with cesium isotopes were investigated in 1988. An analysis of variance of these results showed a significant difference between the four varieties in each type of soil and for each isotope confirming a high root uptake of radiocesium in the barley variety Apex. In 1989 this part of the experiment was continued. The soil in the pots used were ploughed and resowed with the same varieties of barley as used in 1988. The pattern obtained was identical to that in 1988 confirming a high root uptake of Sila and a significantly lower root uptake of Apex and Golf.

In order to confirm the results obtained, the part of the experiment carried out in clay loam was repeated in 1989 with two additional varieties, Gunnar and Ida. Again the same pattern was found, a high root uptake of Sila and significantly lower root uptake of Apex.

The results for barley straw are shown in Table 3.3.2, reported in the same way as for barley grain. As for the grain, an analysis of variance shows, for all categories and year, a significant difference between the varieties. As for the grain, Sila dominates with the highest uptake and Apex with the lowest uptake.

In agreement with earlier experiments (3) the uptake in the straw is somewhat greater than in the grain. In 1988 the ratio grain/straw (Bq/kg per Bq/kg, dry weight) was about 1.5 to 2, in 1989 the ratio has decreased nearly to one. The higher ratio in 1988 might be due to resuspension.

Because of chemical similarity between Cs and K, the Cs uptake from contaminated soil is related to the uptake of K, confer (1). The barley samples have been analyzed for K and no significant differences in the uptake of K between the varieties have been identified.

Between the varieties of barley no significant difference in the yield of dry matter occurred. Consequently the yield was not considered in the discussion.

Table 3.3.1 Root uptake of radiocesium in barley grain given as relative concentration ratios. The results are normalised to the mean in each group (year-soil-isotope category). Each individual value is the mean of four replicates $\pm$ 1SE.

Soil and isotope	Variety	1988	1989
A:			
Organic soil experimentally added Cs-134	Golf	0.87 $\pm$ 10%	0.88 $\pm$ 7%
	Apex	0.86 $\pm$ 7%	0.84 $\pm$ 8%
	Anker	1.10 $\pm$ 7%	1.26 $\pm$ 13%
	Sila	1.17 $\pm$ 7%	1.02 $\pm$ 19%
B:			
Organic soil Chernobyl Cs-137	Golf	0.84 $\pm$ 10%	0.81 $\pm$ 8%
	Apex	0.82 $\pm$ 7%	0.97 $\pm$ 13%
	Anker	1.08 $\pm$ 6%	1.12 $\pm$ 13%
	Sila	1.26 $\pm$ 9%	1.09 $\pm$ 16%
C:			
Clay-loam experimentally added Cs-137	Golf	0.95 $\pm$ 5%	0.94 $\pm$ 5%
	Apex	0.87 $\pm$ 9%	0.67 $\pm$ 10%
	Anker	1.01 $\pm$ 7%	1.02 $\pm$ 4%
	Sila	1.17 $\pm$ 4%	1.37 $\pm$ 7%
Cl:			
Clay loam experimentally added Cs-137 1989	Golf		0.87 $\pm$ 3%
	Apex		0.79 $\pm$ 3%
	Anker		1.08 $\pm$ 12%
	Sila		1.17 $\pm$ 12%
	Gunnar		1.06 $\pm$ 5%
	Ida		1.04 $\pm$ 2%

Table 3.3.2 Root uptake of radiocesium in barley straw given as relative concentration ratios. The results are normalised to the mean in each group (year-soil-isotope category). Each individual value is the mean of four replicates ± 1 SE.

Soil and isotope	Variety	1988	1989
A:			
Organic soil experimentally Added Cs-134	Golf	0.89 $\pm$ 12%	0.88 $\pm$ 14%
	Apex	1.00 $\pm$ 9%	0.88 $\pm$ 5%
	Anker	0.91 $\pm$ 9%	1.09 $\pm$ 11%
	Sila	1.20 $\pm$ 9%	1.18 $\pm$ 9%
B:			
Organic soil Chernobyl Cs-137	Golf	0.92 $\pm$ 12%	0.85 $\pm$ 11%
	Apex	0.84 $\pm$ 11%	0.75 $\pm$ 26%
	Anker	0.92 $\pm$ 8%	1.00 $\pm$ 13%
	Sila	1.32 $\pm$ 9%	1.41 $\pm$ 4%
C:			
Clay-loam experimentally Added Cs-137	Golf	1.04 $\pm$ 5%	1.12 $\pm$ 15%
	Apex	0.82 $\pm$ 8%	0.64 $\pm$ 11%
	Anker	0.83 $\pm$ 10%	0.88 $\pm$ 7%
	Sila	1.32 $\pm$ 7%	1.37 $\pm$ 5%
C1:			
Clay-loam experimentally Added Cs-137	Golf		0.88 $\pm$ 9%
	Apex		0.88 $\pm$ 2%
	Anker		0.88 $\pm$ 9%
	Sila		1.41 $\pm$ 12%
	Gunnar		0.93 $\pm$ 3%
	Ida		1.02 $\pm$ 6%

3.3.2.2.2 Grass

The results for the grass varieties are shown in Table 3.3.3 The concentration ratios are averaged over the growing season for each variety and normalized in the same way as the results for barley.

For all the categories, in 1988 as well as in 1989, the Italian rye grass shows a significantly higher root uptake than the other varieties. This might be partly due to the greater part of connective tissue than the other varieties and a higher yield. To build this connective tissue, the plants use potassium. As stated above the Cs uptake is related to the uptake of K. This might explain the differences in the uptake of Cs between the Italian rye grass and the other varieties.

Italian rye grass is much used in Denmark as pasture grass due to the higher yield in the beginning of the growing season and it is grown only for one year in each field. The qualities of Italian rye grass fit very well with the intensive farming in Denmark where rotation of crops is used as a normal agricultural practice.

The differences between the rye grass varieties during the growing season are shown in Figure 3.3.1. The varieties were sown in 1988 and have continued growing through 1989. The differences between the Italian rye grass and the perennials in 1988 are very clear. The concentrations of cesium in the Italian rye grass are about a factor of 2 to 3 times greater than for the perennials through the whole growing season. These differences decrease in 1989 partly due to plant base absorption (4) which is a dominating factor for the perennials. Thus the cesium concentrations in 1989 for the perennials are the same as in 1988, while the concentrations for Italian rye grass, when used for more than one growing season, reflects the fact that the main part of radiocesium in the clay-loam is firmly fixed already in the first year after the contamination (5).

3.3.3 FIELD STUDY CONCERNING DIRECT CONTAMINATION

In the summer of 1986 a number of barley varieties were harvested at the Danish state experimental farms and analyzed for radiocesium. These farms are located in different areas of the country, which means that the barley varieties have been exposed to very different amounts of radiocesium deposition after the Chernobyl accident. According to Aarkrog et al. (6) the total deposition of Cs-137 in Denmark ranged from a low of about 800 Bq m⁻² in Zealand to a high of about 3600 Bq m⁻² in the western part of Jutland. In addition the locations of the state experimental farms comprise different soil types and climatic conditions of the country thus providing different growing conditions for the crops.

Table 3.3.3 Root uptake of radiocesium in rye grass given as relative concentration ratios, averaged over the growing season. The results are normalised to the mean in each group (year-soil-isotope category). Each individual value is the mean of four replicates $\pm 1\text{SE}$.

Soil and isotopes	Variety	1988	1989
A:			
Organic soil	Italian	1.50 $\pm$ 13%	1.29 $\pm$ 14%
experimentally	Early		
added	Perennial	0.78 $\pm$ 21%	0.92 $\pm$ 11%
Cs-134	Late		
	Perennial	0.72 $\pm$ 16%	0.79 $\pm$ 12%
B:			
Organic soil	Italian	1.43 $\pm$ 19%	1.32 $\pm$ 31%
Chernobyl	Early		
Cs-137	Perennial	0.82 $\pm$ 30%	0.84 $\pm$ 25%
	Late		
	Perennial	0.75 $\pm$ 26%	0.84 $\pm$ 25%
C:			
Clay-loam	Italian	1.69 $\pm$ 21%	1.29 $\pm$ 10%
experimentally	Early		
added	Perennial	0.62 $\pm$ 19%	0.85 $\pm$ 17%
Cs-137	Late		
	Perennial	0.70 $\pm$ 25%	0.85 $\pm$ 17%
Cl:			
Clay-loam	Italian		1.24 $\pm$ 13%
experimentally	Early		
added Cs-137 1989	Perennial		0.76 $\pm$ 16%

In spite of these differences in the deposition pattern, which is reflected in the cesium concentrations of the grain, the results obtained from each experimental farm show distinct similarities. The results for the winter barley varieties are shown in Table 3.3.4. An analysis of variance shows significant differences between the varieties: the Hasso variety and the variety WW 1261 are the most sensitive varieties to direct contamination and AC 456/10 is the least sensitive variety. The analysis of variance shows no significant interaction between varieties and locations, which confirms that the pattern between the different varieties are independent of the locations despite the different levels of deposition and different growing conditions.

The results for spring barley are shown in Table 3.3.5. As for winter barley an analysis of variance shows significant differences between the cesium concentrations of the different varieties and no interaction between varieties and locations.

The fact the cesium concentrations in the winter barley exceed the cesium concentrations in the spring barley reflects the time of the contamination in the early spring, where winter barley has emerged but not spring barley. This means that the main part of the contamination of spring barley is due to direct contamination of radiocesium resuspended from the surface soil.

3.3.4 COUNTERMEASURES

3.3.4.1 Countermeasures concerning indirect contamination

As shown in 3.3.3.2.1 we have identified a barley variety Sila with a high root uptake and a barley variety Apex with a significantly lower root uptake of radiocesium both in clay-loam and organic soil.

Similarly, for the grass varieties the Italian rye grass was identified among the species tested with a relatively high uptake of radiocesium in both the organic soil and in the clay-loam.

It has been possible to identify these differences in two very different types of soil and different types of crops. This indicates that it might be possible, for many types of crops grown in a given type of soil, ranging from clay-loam to organic soil, to identify a variety with a low radiocesium contamination potential.

In order to demonstrate quantitatively the implications of these results, we can look at a few examples:

In order to compare the uptake of different crop species with different yields, we may introduce the common unit activity per fodder unit, Bq/FU. The results for crops grown in clay-loam are shown in Table 3.3.6.

One can see that for a given contamination the concentration in barley grain grown in clay-loam is a factor four to six times smaller than in the straw for all varieties and up to nine times smaller than in the grass. That means if it was possible for the farmer to change species without regard to fodder plans for the domestic animals, he should prefer barley grain.

Table 3.3.4 Concentration of directly Cs-137 from Chernobyl in winter barley in Denmark, 1986, (Bq/kg fresh weight) relative units (fresh weight, the mean in each group equals to one the relative scale).

Location	Variety	Relative units	Mean, Bq/kg fresh weight
Godthab	Igri	0.86	0.891
	Gerbelt	0.72	
	Hasso	1.37	
	AC 456/10	0.67	
	WW 1261	1.36	
Borris	Igri	0.70	1.82
	Gerbelt	0.64	
	Hasso	1.27	
	AC 456/10	0.66	
	WW 1261	1.72	
Ronhave	Igri	0.91	3.33
	Gerbelt	0.96	
	Hasso	1.21	
	AC 456/10	0.88	
	WW 1261	1.04	
Jullerup	Igri	0.87	0.897
	Gerbelt	0.81	
	Hasso	1.37	
	AC 456/10	0.63	
	WW 1261	1.25	
Karise	Igri	0.91	0.892
	Gerbelt	0.82	
	Hasso	1.38	
	AC 456/10	0.64	
	WW 1261	1.25	
Roskilde	Igri	0.81	0.665
	Gerbelt	0.70	
	Hasso	1.51	
	AC 456/10	0.80	
	WW 1261	1.18	

Table 3.3.5 Concentration of directly deposited Cs-137 from Chernobyl in spring barley in Denmark 1986 (Bq/kg fresh weight) relative units (the mean in each group equals to one on the relative scale).

Location	Variety	Relative units	Actual mean Bq/kg fresh weight
Tylstrup	Anker	1.02	0.215
	Claro	1.24	
	Sila	0.77	
	WW 7200	0.96	
Godthab	Anker	1.74	0.270
	Claro	1.11	
	Sila	0.80	
	WW 7200	0.76	
Grindsted	Anker	1.00	0.672
	Claro	1.09	
	Sila	0.92	
	WW 7200	0.98	
Tystofte	Anker	1.02	0.142
	Claro	0.98	
	Sila	0.94	
	WW 7200	1.05	
Abed	Anker	1.41	0.242
	Claro	1.30	
	Sila	0.03	
	WW 7200	0.05	

Table 3.3.6 Activity retained in the crops, grown in clay-loam, expressed in Bequerels per Fodder Unit (Bq/FU). Average of four replicates ± 1 SE

Species	Variety	Grain	Straw
Barley	Golf	10000 $\pm$ 9%	63900 $\pm$ 9%
	Apex	9000 $\pm$ 3%	64000 $\pm$ 2%
	Anker	12400 $\pm$ 7%	63500 $\pm$ 9%
	Sila	13500 $\pm$ 12%	102300 $\pm$ 12%
	Ida	12000 $\pm$ 2%	67100 $\pm$ 4%
	Gunnar	12300 $\pm$ 5%	73900 $\pm$ 7%
Grass	Italian		91200 $\pm$ 3%
	Early perennial		33800 $\pm$ 24%
	Late perennial		44800 $\pm$ 6%

* 1FU is the fodder value of 1 kg. of barley

In Denmark the main part of the fodder for pigs consist of barley grain. The results in Table 3.3.6 show that a change of variety from Sila to Apex reduces the activity per fodder unit, and thereby the activity in pork used for human consumption, by 30% after a given contamination.

The transfer of cesium decreases with about a factor of 2 (7) when the fodder contains fibrous components. However a shift from fodder with a high grain ratio to fodder with a high hay ratio, will raise the activity in the animal produce, due to the higher concentration of cesium in the straw.

An even more pronounced effect of varieties is seen for the pasture grass, were the activity per fodder unit in the Italian rye grass is a factor of 2 to 3 times higher than for the other varieties. Reducing the activity per fodder unit by a factor two or three by choosing another rye grass variety, will reduce the activity in the animal produce (milk and beef) correspondingly.

In summary, we can state that it is possible to reduce the radiological consequences after a nuclear accident by efficient agricultural planning of countermeasures, comprising changes of agricultural practices towards the use of plant species and varieties with low root uptakes. The countermeasures can contribute to a significant reduction of the contamination levels perhaps even below a critical level.

It is worth noticing that this kind of countermeasures costs nothing for the farmers and has no adverse side effects like pollution, e.g. compared with countermeasures like fertilizing with potassium, sodium etc.

3.3.4.2 Countermeasures concerning direct contamination

The results stated in section 3.3.3 mean that interception by and translocation in the barley varieties after a direct contamination with radiocesium can be reduced if the farmers choose varieties which have low sensitivities for direct contamination. However due to the psychological implications such prophylactic countermeasures are not realistic in the context of nuclear accidents.

The results could be of importance for agricultural areas close to nuclear power plants with routine discharges to the atmosphere. Without any additional costs, by proper planning of the agricultural practice, it is possible to reduce the contamination of animal produce from this area. This might also be considered after an accident, in areas with high resuspension.

3.3.4 CONCLUSION

Different crop species and varieties have been tested for their sensitivity to root uptake of radiocesium from different types of soil. Significant differences have been revealed and the results have been discussed with regard to countermeasures after a nuclear accident.

Furthermore different barley varieties have been tested for their sensitivity to direct contamination. As for the indirect contamination, significant differences have been identified and discussed with regard to countermeasures.

In summary, we can state that efficient agricultural planning of countermeasures to reduce the radiological consequences after a nuclear accident can contribute to a significant reduction of the contamination levels perhaps even below a critical level.

The kind of countermeasures recommended cost nothing for the farmers and have no side effects like pollution, e.g. compared with countermeasures including fertilizing with potassium, sodium etc.

The general pattern of the results indicate that they are to be found for varieties and soil types other than those investigated. The results could thus prove to be of general value to all member states of the EEC.

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4. DAIRY INDUSTRY

4.1 RADIOCESIUM TRANSFER FROM WHOLE MILK TO A RANGE OF MILK PRODUCTS

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4.1.1 INTRODUCTION

Milk, a white coloured fluid of low viscosity and slightly sweetish taste, is a complex mixture of proteins, fats, carbohydrates, salts and water. Some of its constituents such as milk-fat, sugar and protein are not found elsewhere, either in the body or in nature. The quantity of the main constituents of milk can vary considerably between cows of different breeds and individual cows of the same breed. Therefore, only limit values can be stated for the variation. Typical values for milk composition are given in Table 4.1.1 The dairy industry is unique in producing from a single raw material a complex range of products. Nowadays, variations in the composition of milk from one country to another are usually quite small. International Standards proposed by the International Dairy Federation, the Food and Agriculture Organization of the United Nations, and the World Health Organization have done much to standardize dairy products throughout the world.

Milk is a perishable raw material which must be stabilized against bacterial spoilage and which can be converted into consumable products through a multiplicity of processing procedures. Pasteurization and more rigorous heat treatments, such as sterilization, are required for microbiological control in the interest of public health. Other processes, such as homogenization, concentration, dehydration, freezing, renneting and churning, are employed to modify the composition of milk en-route to the production of the myriad of dairy products available to the consumer.

Radioactivity of milk transferred through the food chain via forage and water intake by ruminants is a good indicator of the degree of environmental contamination post accident. Milk and its products constitute a substantial portion of the human diet and represent one of the principal means by which food-borne radionuclides are ingested. The Chernobyl accident and subsequent widespread contamination demonstrated clearly that the dairy industry is highly sensitive to air-borne pollution. Consequently, much attention has been given to possible ways of limiting the transfer of environmental contaminants to man via milk. Three methods of approach offer themselves: (a) prevention or reduction of the entry of radioactivity into milk; (b) identifying those milk products with least contamination with a view to

expanding their production; and (c) removal of radionuclides from milk by appropriate ion exchange techniques.

The second option was addressed in detail in this study whose overall objectives were as follows:

- 1) To conduct a thorough and systematic search of the published literature with a view to:
 - (i) compiling a comprehensive bibliography of relevant publications;
 - (ii) carrying out a critical review of this corpus of knowledge; and
 - (iii) identifying shortcomings in our present understanding and making recommendations regarding possible future research.
- 2) To carry out detailed field studies of the transfer of radiocaesium to dairy products following the processing of contaminated whole milk with particular emphasis on transfer coefficients; and
- 3) To formulate guide-lines for the dairy industry which will enable the industry and Public Health Authorities to respond promptly in the event of contamination following a nuclear accident.

4.1.2 LITERATURE REVIEW

There has been considerable interest over the years in the possibility that contaminated milk can be processed into a less contaminated food with a consequent reduction in the radiation doses received by consumers. Published reports date back to the sixties following research conducted in the aftermath of the atmospheric weapons testing programmes of the late 1950s and early 1960s. Since then few data have been published until recently, when there has been renewed interest due mainly to the Chernobyl accident. Few attempts have been made to review this literature [2, 3, 4].

The comparability of data is hindered by the variety of definitions used in the literature to define changes in the radionuclide content of food [2]. In the case of milk the situation is not as acute as only two definitions have customarily been used, namely:

- a) the total amount of a radionuclide in processed food divided by the total amount of this radionuclide in the original raw food. Known as F_p , this fraction of the radionuclide which remains in the food after processing is often expressed as a percentage; and
- b) The ratio of the radionuclide concentration as measured in the raw and the processed product.

The F_p or % transfer value is thought to be the best way to define changes in radionuclide content because (i) much literature data is already defined in this way, and (ii) conversion to F_p values appears to be possible for nearly all collected literature data.

In most of the early studies the milk was obtained from animals artificially fed on radioactive materials and the products were prepared on a small-scale batch basis in the laboratory. Few studies were carried out on samples containing environmental levels of radioactivity which had been obtained from commercial milk processing plants.

Radiostrontium, radioiodine and radiocaesium are amongst the few radioelements whose behaviour under dairy processing has been studied to a significant extent. Indeed, in the early work more emphasis was given to radiostrontium than radiocaesium. Post-Chernobyl research has focussed mainly

on iodine-131 and radiocaesium.

As early as 1957, Booker [5] measured the radiocaesium and radiostrontium levels in dried milk. In this paper the method used for measuring 'fallout' caesium-137 and natural potassium is described and the caesium-137 concentrations observed during the period 1954-1956 are compared to the strontium-90 concentrations in the same samples.

The method chosen to determine caesium-137 and potassium-40 activity was by gamma analysis using a 3" x 3" NaI(Tl) crystal and five channel analyser. Skimmed dried milk samples dating from 1953 to 1957 were analysed. Potassium levels were found not to vary significantly between samples but the caesium-137 activity showed a sharp increase by a factor of 3. A similar increase was noted for the strontium-90 content of milk [6], and was associated with the observed sharp increase in the rate of fallout in May 1955 [7]. The increase may also have been related to the fact that at about the same time cattle normally change from winter feed (harvested in the previous summer) to 'contaminated' summer feed. A similar, but less marked, step was detected in 1956.

In regard to studies conducted to investigate the transfer of radionuclides to milk products in the course of processing, one of the earliest was in 1960 by Demott and Cragle [8], who examined the distribution of strontium-89 and calcium-45 in skimmilk, cream, Cheddar cheese and whey from milk obtained at various times following ingestion of these nuclides by Jersey cows. They also looked at how this distribution compared for products made from dosed milk as opposed to dosed cows. In the former case they found that the concentrations of these nuclides in cream (40% fat) were approximately half those in the whole milk from which it was separated, whilst the concentrations in skimmilk were slightly higher. The concentrations in the cream separated from dosed milk were also halved. They further observed that cream containing strontium-89, after repeated washings by dilution and reseparation, resulted in a product essentially free of this nuclide. Variations in strontium-89 concentration in cream after separation were greater for cream from dosed cows. This can be partially explained on the basis of the time lapse between dosing of the cow and the separation process.

Cheddar cheese produced from milk containing strontium-89 and calcium-45 showed a slightly larger ratio of strontium-89/calcium-45 than did the milk from which it was made. Fourteen vats of cheese made from dosed milk were examined and shown to have a mean

$$\frac{\text{strontium-89/calcium-45 (cheese)}}{\text{strontium-89/calcium-45 (milk)}}$$

ratio of 1.23 ± 0.28 , whilst four vats of cheese made from the milk of dosed cows showed a ratio of 1.06 ± 0.12 . The strontium-89/calcium-45 ratio in whey was less than in the milk. These results indicate that cheese contains more strontium per gram of calcium than does the milk from which it is made, a point worth noting if considering the manufacture of cheddar cheese from milk contaminated by strontium.

Lengemann [9] studied the distribution of strontium-85 and caesium-137 in a range of common dairy products produced from milk obtained from Holstein cows being given daily oral doses of these nuclides. The milks were divided into whole milk, skimmilk and cream. The cream was then used to produce an unsalted sour cream butter, the whole milk an unwashed cheddar curd and the skimmilk a washed cottage cheese curd. Lengemann also quoted the weight of the materials and the moisture content of the products so that a comparison can be made between these laboratory products and their commercial counterparts. He observed that about 93% and 7% of the strontium-85 activity present in the whole milk was transferred to the skimmilk and the cream, respectively. The figures for caesium-137 were almost identical at 94% and 6%, respectively. Soxhlet extraction of the cream indicated that no radioactivity was associated directly with the fat and explained the low degree of transfer from whole milk to butter. All researchers appear to be agreed on this point. Indeed, Assimakopoulos *et al.* [10, 11], in a comparative study of the transfer of iodine-131 and radiocaesium from sheep's milk to cheese (Gruyère) products following the Chernobyl accident, demonstrated that caesium showed a preferential concentration in aqueous by-products, doubtless, due to its considerably greater solubility in aqueous media. To further emphasize this point Lengemann found that 3.6% of the radiostrontium in cream was recovered in the butter. This translates into a 0.6% transfer from the whole milk. Similarly with the radiocaesium, 5.1% of the activity was transferred from the cream, which amounts to 0.3% of the

activity in the whole milk.

The transfer of strontium-85 from whole milk to cottage cheese, at 2.7%, was much less than for the cheddar cheese (49%). This is not surprising given the relative calcium contents of the two types of cheese and the fact that Demott and Cragle [8] demonstrated that only minor differences exist in the behaviour of calcium and strontium during cheese manufacture. Much less caesium than strontium was recovered in the cheddar cheese (7.7%), which probably reflects the degree of association of these nuclides with the protein. Also, only a very small fraction (1.1%) of the caesium was found in the cottage cheese.

Similar results to those obtained by Demott and Cragle [8] and Lengemann [9] for radiostrontium were arrived at by Buma and Meerstra [12], who studied the transfer of fallout strontium-89 and strontium-90 from raw milk to Gouda cheese and whey. Buma and Meerstra concluded (from the results obtained by Demott and Cragle, and Lengemann) that the manner in which the strontium-tracer is introduced into the milk may influence the transfer of this isotope from the milk to the cheese. Accordingly, in order to study the movement of fission products through the food-chain, they considered that it was preferable to measure the 'fallout' strontium activity in milk and its products. In a period of relatively high strontium radioactivity, weekly samples of raw milk and the curd, whey and cheese (Gouda) made from it, were taken from a cheese factory and measured for radiostrontium. The mean value of the

$$\frac{\text{strontium/g calcium in (cheese)}}{\text{strontium/g calcium in (milk)}}$$

ratio was 1.18 ± 0.08 , whilst the corresponding whey/milk ratio was 0.68 ± 0.04 . Both values are in close agreement with those found by Demott and Cragle for cheese made from dosed milk.

Dubrovina and Belova [13] examined the relative transfer of calcium, stable strontium and strontium-90 from bovine milk during standard processing to a wide range of dairy products including cream, skim milk, casein, butter, buttermilk and cottage cheese. They found that the relative content of strontium-90 per gramme of calcium and per milligramme of stable strontium

was practically the same in all these products and that only 4 - 5% of the strontium-90 passed from milk to cream and less than 1% to butter, in agreement with Lengemann's results. The absolute content of strontium was not uniform in the various products, it was highest in casein - the protein fraction of milk - and lowest in cream. In terms of percent transfer from whole milk, between 20 - 25% of strontium and calcium were recovered in the casein and 60 - 70% in buttermilk. Ohmomo and Tsugo [14] reported that when whole milk was separated into skimmilk and cream about 90% of added strontium-89 and caesium-134 were transferred to the skimmilk component. Washing was found to remove both nuclides from the cream. Little or no radioactivity was detected in the fat fraction of the butter, whilst most of the caesium-134 ended up in the whey fraction. Approximately 45% of the strontium-89 in the original milk was found in Gouda-type cheese and proved quite difficult to remove.

Calapaj and Ongaro [15] carried out laboratory and semi-industrial investigations on the passage of strontium-90 and caesium-137 from fallout contaminated milk to butter and cheese with results which confirmed previous studies. Both strontium and caesium were present in butter to less than 1.5%. On the other hand, in rennet coagulation the strontium-90 in the coagulum ranged from 80% to about 20% of that present in the milk and decreased with increasing acidity in a similar fashion to calcium. The caesium-137 in coagulum was always less than 10% and was independent of the acidity.

Kirchmann *et al.* [16] evaluated the possibilities of using milk products made from milk contaminated with strontium-85, iodine-131 and caesium-134 at levels in excess of the standards prescribed for direct human consumption and, like Demott and Cragle [8], examined whether the distribution of these nuclides was identical in products made from milk contaminated *in vivo* and *in vitro*. The products studied were manufactured on a laboratory scale geared to the technological processes used in the dairy industry. The mode of contamination was found to have little influence on the distribution of these nuclides in skimmilk, cream, butter and buttermilk with the exception of iodine-131 in butter, which differed by a factor of at least 2 depending upon the mode of contamination. Furthermore, radiostrontium contamination of cheese-type products made from milk contaminated *in vivo* was very strongly

influenced by the mode of coagulation, whereas it would appear to be of little importance in the case of milk contaminated *in vitro*. Kirchmann also compared the uptake of radionuclides in the various products and examined the possibility of using milk contaminated by fission products with particular emphasis on the effect of technological processing on the distribution of the radionuclides in certain products. This last point was also investigated by Dubrovina and Belova [13], Calapaj and Ongaro [15], and Micic *et al.* [17, 18]. The former two recommend acid coagulation for cheese production as the strontium concentration in the coagulum decreased with increasing acidity. The latter demonstrated that biochemical composition and technological procedures in milk processing can contribute to a reduction in potassium-40 levels in milk products and, by extension, caesium-137 levels.

Lagoni *et al.* [19, 20], in a survey of the distribution of weapons fallout strontium-90, iodine-131 and caesium-137 in milk products, reported that strontium and caesium were only detected in the non-fat constituents of milk, whereas part of the radioiodine was also present in the milk fat. They measured the strontium-90 levels in acid and rennet casein and obtained a transfer from the whole milk of 6.3% and 84.7%, respectively, confirming the results of previous studies. The distribution of caesium and strontium across the products were found to be similar except for casein and cheese. Iodine, on the other hand, though behaving in a similar fashion to caesium, is present in butterfat (2.1%). The mode of coagulation for casein production appears to affect the iodine distribution (3.9% and 2.0 % for acid and rennet casein, respectively), though the difference is not as marked as for strontium.

Reavey [21, 22] investigated the distribution of strontium-90, iodine-131 and caesium-137 in samples of whole milk and milk products obtained from two processing plants serving the Boston area in the U.S. The levels of the radionuclides observed were those present in the environment due to weapons testing fallout. Under these conditions the behaviour of the radionuclides would be representative of actual field conditions, unlike most previous studies where the milk was obtained from cows fed on contaminated feed and the products then manufactured on a laboratory scale. Reavey, in accordance with previous studies, observed that the relative distributions of

strontium-90 and caesium-137 in milk and milk products were similar, with cottage cheese, heavy cream and butter retaining by far the smallest concentrations. He also investigated the stable calcium and potassium content and found that the strontium-90 to stable calcium ratio and caesium-137 to stable potassium ratio showed little variation and concluded that strontium-90 follows calcium and caesium-137 follows potassium in each of the separation processes examined.

Reavey also looked at the distribution of iodine-131 in milk products. He observed that in the cream, in contrast to caesium and strontium, the specific activity of iodine-131 was twice that of the original whole milk. This translates into a 20% transfer (cream is 10% by weight of whole milk), as opposed to an approximate 6% transfer for caesium-137. The iodine-131 concentration in evaporated skim milk (a concentrate containing 40% milk solids) was about 25% greater than that in the original whole milk. However, given the weight loss by evaporation, ~80%, the iodine-131 content amounts to only about 30% of that in the original sample. This indicates a direct loss of iodine-131 in the evaporation process. Continued evaporation to produce non-fat dry milk reduced the iodine-131 content still further, to 8%. This finding is in disagreement with that of Raymond and Williams [23], who reported no loss of iodine-131 content in the preparation of evaporated whole milk and only a 40% loss in preparation of non-fat dry milk. Reavey suggested that this discrepancy could have been due to the different manufacturing processes employed.

In 1984, Kerkhof Mogot *et al.* [24] investigated the extent of iodine-131 losses during the processing of contaminated whole milk into milk powder. Unlike previous researchers [21, 22, 23], who obtained milk from milk processing plants, Kerkhof Mogot *et al.* used milk from a cow to which a known quantity of iodine-131 was administered orally on each of 4 successive days. The milk was pasteurized, concentrated by evaporation and spray dried. Samples of milk taken before and after pasteurization were analysed for radioactivity content, as were samples of the milk concentrate and distillate to determine any possible losses of radioiodine.

The results of the study demonstrated that in the manufacture of whole milk

powder no loss of radioactive iodine in volatile form occurred, neither during pasteurization and evaporation of the milk, nor during drying of the concentrate in the spray drier. This conclusion was based on the observation that throughout the whole process the radioactivity per kg of dry matter was almost constant. With regard to the evaporation process, this conclusion is confirmed by the absence of radioactivity in the distillate. This result is in reasonable agreement with that obtained by Raymond and Williams who found some loss of radioactivity in drying. This loss could possibly be attributed to the fact that in the drying tower the powder may not be completely trapped in the cyclone and a proportion of the powder may be left behind on the wall of the drier. With regard to the pasteurization step, Demott and Easterly [25] have shown that iodine-131 is not volatile when milk is heated to 70°C in a vacuum.

Mayes White and Moghissi [26], in order to evaluate the potential radiation exposure of man, studied the transfer of iodine-131 from *in vivo* and *in vitro* labelled cows' and goat' milk to cheese. Like all previous studies on the transfer of radionuclides to cheese, the results displayed a strong dependence on the type of cheese manufactured [16]. Mayes White and Moghissi observed that for *in vivo* labelled milk the percent transfer was 7.9% for goats milk (80g cheese/litre of milk), 11% for milk from a Holstein cow and 17.7% from a Jersey cow. The corresponding results for the *in vitro* labelled milk were 6.2% (66g cheese/litre of milk), 11% and 15%. Evidently the mode of contamination does not greatly influence iodine distribution. However storage time of the milk before cheese manufacture does, with increased transfer occurring for a longer storage time.

Little was published after 1965 until the Chernobyl accident when Kandarakis and Anifantakis [27] examined the distribution of iodine-131, caesium-134 and caesium-137 in products produced from ewes milk by different processes. The percentage transfer of the three nuclides from the raw milk to the products were found to be 4%, 3% and 3%, respectively, in the case of cream, 17%, 10% and 9% in cheese, 5%, 4% and 4% in myzithra, and 45%, 38% and 34% in strained yogurt.

Other studies, carried out in this period by Everitt and Paulsson [28],

Assimakopoulos *et al.* [10, 11], Antila *et al.* [29], Kankare *et al.* [30, 31] and Pirhonen *et al.* [32], confirmed that caesium is mainly found in the non-fat constituents of milk, while some of the iodine is also present in milk fat. In cheesemaking, 5 - 10% of the caesium is transported to cheese and the rest to whey [28, 29]. Assimakopoulos *et al.* [10, 11] highlighted an interesting feature regarding the ripening time of cheese during which period there occurs a 30% loss in weight (presumably dehydration) resulting in an increase in specific radioactivity. It is important to appreciate that Assimakopoulos' study was conducted on sheep' milk and Gruyère cheese, unlike previous studies which examined cows' milk and cheddar/cottage cheese. Furthermore the "cream" referred to was separated from the whey rather than from the whole milk. This would account for the very low percentage transfer of approximately 0.3% compared to about 5% for the normal separation in cows' milk. However, the percent transfer of caesium to cheese is in good agreement with that obtained by Lengemann for cheddar cheese produced from cows' milk.

Wilson *et al.* [33] and Wood *et al.* [34] investigated the transfer of radiocaesium from milk collected in Cumbria, U.K. to a range of commercially available dairy products produced on a pilot/laboratory scale. Their data showed clearly that radiocaesium concentrates in the aqueous fractions during separation and thus high fat fractions and high protein fractions are relatively depleted in radiocaesium. Furthermore, it was confirmed that caesium is comprehensively removed from whey during ion-exchange demineralization. They demonstrated that from a commercial viewpoint, the products containing least radioactivity on an 'as sold' basis are the cheeses, high fat products and demineralized whey powders.

Small differences in actual percentage values for the transfer of a particular nuclide can be accounted for by differences in the methods of processing and the composition of the milk itself. Although the processing schemes are, in principle, similar, no two creameries conduct a process in exactly the same way. This is the case particularly for cheese production which varies considerably in the different creameries. The composition of the milk will also have a bearing on transfer ratios, e.g., the fat content of the cream is known to affect the amount of activity that is transferred.

As caesium favours the aqueous fractions, it follows that the higher the fat content of the cream the lower the activity transferred from the whole milk to the cream.

Pappas *et al.* [35] adopted a slightly different approach and investigated the transfer of radiocaesium from ovine milk to feta cheese in a series of experiments in which modifications were made to the standard cheesemaking procedure. Not surprisingly, they found that the percentage transfer of radiocaesium during feta cheesemaking was practically independent of the factors entering the production stage, e.g., percentage of culture employed and coagulation temperature of milk. Arising from this, their subsequent experiments focused on the possibility of a reduction in radiocaesium concentration during the storage phase. The experiment involved periodic replacements of the storage medium (brine). With each replacement of the brine both the cheese and the removed liquid were measured for radiocaesium content. A reduction of caesium-137 concentration was observed and the results satisfactorily validated the predictions of the simple two-compartment model simulating the diffusion of an ideal gas through a bi-directional membrane. The final product was examined with regard to alterations in composition and palatability. Though some changes did occur, the product in all cases was found to be acceptable.

Further published work pertaining to radionuclides in milk and milk products, though not addressing directly the transfer of radioactivity during processing, will be addressed in Section 4 of this report.

Data, from the studies reviewed above, on the percentage distribution of radiocaesium in a wide range of dairy products, are summarized in Tb. 4.1.2. It is evident from an examination of this table that there are gaps in our knowledge regarding the transfer of caesium to certain milk products.

Although the percentage transfer to cream and skimmilk and then to butter is well established, few studies have been conducted on the transfer of radionuclides to whey and its derivatives, a point also brought out by Giese [36]. This is not surprising given that in the past whey was usually considered a waste by-product and it is only in recent years that whey and

its derivatives have been manufactured on a commercial scale. The few data that are available indicate that approximately 90% of the activity ends up in the whey fraction.

4.1.3 METHODOLOGY

The flow diagram in Fig. 4.1.1 illustrates the principal milk processing pathways. This is typical for a large European creamery. The end products in this diagram are, for the most part, those produced in the greatest quantities. Many of these end products are in turn often used as ingredients for other dairy products and foodstuffs in general (Tb. 4.1.3). The importance of understanding, in a quantitative way, how radiocaesium (and indeed other fission and activation products) are transferred from whole milk to an extensive range of milk products is therefore evident.

Having identified the principal milk processing pathways and the major dairy products, the choice remained as to which creameries would be the most suitable from which to obtain samples. Three of the largest Irish producers were chosen principally because of their wide range of products. A systematic method of sampling was devised which incorporated all the principal processing pathways.

The following is a brief outline of the pathways studied:

STARTING PRODUCT	---	END PRODUCT
(A) Whole milk	---	Whole milk powder
(B) Whole milk	---	Cheese + Cheese whey
(C) Whole milk	---	Skimmilk + Cream
(D) Skimmilk	---	Skimmilk powder
(E) Skimmilk	---	Casein + Casein whey
(F) Cream	---	Butter/Butteroil +Buttermilk
(G) Whey	---	Whey powder
(H) Whey	---	Lactose + Delactosed whey powder
(I) Whey	---	Demineralized whey (Demin whey)
(J) Whey	---	Lactose + Partially demineralized delactosed whey (Demindelac)
(K) Demin whey	---	Demin whey powder

Sampling of the above pathways, facilitates the determination of

radiocaesium transfer from whole milk to any dairy product. For example, the transfer behaviour of caesium from whole milk through to lactose can be determined by examining paths B, C + E and H (Fig. 4.1.2).

The milk production cycle in Ireland, as indicated by the percentage monthly milk intake by creameries, is given in Fig. 4.1.3 and highlights the untimeliness of the Chernobyl accident, occurring as it did at the beginning of peak production. In the aftermath of the Chernobyl accident, radiocaesium levels in Irish milk declined steadily from approximately 200 Bq/kg in early May to about 3 Bq/kg by late October 1986 [37, 38]. By December 1986 mean levels had risen to about 10 Bq/kg, as farmers reverted to winter feed harvested the previous summer when levels of contamination were higher.

Given the distinct pattern of production throughout the year, it was decided to initiate sampling at the onset of peak production and to continue sampling periodically to the end of the year, by which time many creameries cease production of milk products. In this study, a total of 400 samples of milk and milk products were taken at three creameries over identical seven-month periods in 1988 and 1989. The timing and the number of samples collected from each creamery is given in Table 4.1.4

Two high resolution gamma spectrometers were employed in the present work. The first, a lithium-drifted germanium detector linked to a Silena 8K MCA equipped with digital spectrum stabilisation and interfaced to a VAX 11/785 computer, and the second, an ultra-low background intrinsic n-type germanium detector linked to a P/C based MCA. Both detectors were heavily shielded with aged lead and spectra were analysed using modified versions of the gamma spectra analysis programme SAMPO80 (39). Before any measurements were conducted and specific activities determined, an efficiency calibration of each detector was performed using a certified multi-line source whose emission rates per gram of solution were known. In this study the standard used was the Reference Material SRM-4276, supplied by the National Institute of Standards and Technology (NIST). It is composed of Sb-125 + Te-125m, Eu-154 and Eu-155 in solution and provides 19 useful calibration points between 27 keV and 1596 keV. This solution was used to prepare laboratory standards identical in geometry and density to the samples being analysed.

The calibration spectra were recorded for 24 hours and efficiency functions determined for each of the different geometries used.

Samples were measured for periods ranging from 1 - 4 days depending on the activity. A typical spectrum is shown in Fig. 4.1.4. Reliability and traceability to the NIST, was established by the use of the above mentioned mixed-radionuclide standard (SRM 4276). The laboratory also participated in a number of international intercomparison exercises. The one most relevant to this project was the Asfalec ring test on the radioactivity content in dried milk powder, organised in 1987 by the Netherlands Controlling Authority for Milk and Milk Products. Satisfactory agreement, within the overall uncertainties, was achieved in this test.

4.1.4 RESULTS & DISCUSSION

The results of the analyses of the various pathways considered in this study are presented in Tb. 4.1.5-4.1.25. The caesium-137 concentrations in whole milk, though low, were relatively uniform throughout most of the study period, with values ranging from 0.2 Bq/l to about 1.1 Bq/l (only one sample measured greater than 1 Bq/l). By way of comparison, prior to the Chernobyl accident, the mean caesium-137 concentration in Irish whole milk was 0.09 Bq/l [40].

The near steady-state situation which prevailed was not without advantage in the determination, under commercial/industrial rather than controlled laboratory conditions, of representative transfer factors. The calculation of the percentage transfer of a particular radionuclide from starting material to end product is straightforward, requiring only the concentration of the radionuclide in both materials together with the yield of the relevant end products, i.e., the quantity of starting material required to produce 1 kg of end product. The latter can and does vary from creamery to creamery depending upon the particular processing method employed and the composition of milk (Tb. 4.1.1), which can change considerably according to season and weather (Figs. 4.1.5 & 4.1.6).

The distribution of caesium-137 in the different products analysed in the present study, expressed as a percentage of the activity in the starting material of the particular pathway, is presented in Table 4.1.26. The percentage activity transferred from whole milk to each product is presented in Fig. 4.1.7 and detailed in Tb. 4.1.27 & 4.1.28. It is evident from these data that the behaviour of caesium-137 conforms closely to expectation with the bulk of the activity following the skim fraction, leaving the fat fraction relatively free. The measurements indicate that, on average, 93% of the activity remains in the skimmilk, well within the range of 85 - 99% observed in previous studies [9, 14, 19, 20, 21, 22, 29, 30, 33, 34]. Wilson *et al.* [33] and others [10, 11, 27, 28, 29, 30, 31] have demonstrated that caesium, a highly soluble monovalent cation, has a strong tendency to concentrate in the more aqueous fractions during physical separation, whilst the fat fractions and high protein fractions are relatively depleted in caesium. The data from this study is in excellent agreement with these findings; for example, the

percentage transfers to cream and cheddar cheese are 6.5% and 6.9% , respectively.

The percentage activity transferred to cream depends on its fat content, which explains the wide range of values found in the literature (3 - 16%). In this study, the fat content of the cream was typically 40%.

The caesium-137 concentration in cheese depends significantly on the type of cheese made and the processing method used. Kirchmann [16] demonstrated this clearly by measuring the activity transferred to a wide variety of cheeses; hard, soft, acid or rennet. This is reflected in the wide range of values reported in the literature. In the present study, an average transfer of 6.9% was observed. In certain cases the activities measured were below detection limits and could not, therefore, be used to determine the percentage distribution. Overall, 91% of the caesium was found in the cheese whey.

The bulk of the activity that remains in the skimmilk is transferred to the casein whey (~83%). The levels of activity in the casein itself were so low that only MDA's could be reported. The highest obtained, <2.6%, which translates into <2.8% of the activity from whole milk, falls within the range found by others (1.8 - 7.7%) [16, 19, 20, 30, 31].

As with the casein, a similar problem was encountered with certain butter samples: no activity could be detected and consequently only MDA's could be determined. Approximately 66% of the activity in the cream ends up in the buttermilk, which is equivalent to 4.3% of the activity from the original whole milk. Assuming no loss of caesium (as there is no waste product), it can be inferred that about 34% of the activity goes to the butter, which amounts to 2.2% of the activity in the whole milk. These values are in agreement with those reported in the literature for butter (0.3 - 2.2%) and buttermilk (2 - 13%), (Tables 4.1.2 & 4.1.28).

A small number of butteroil samples were also analysed. Since butteroil is 99.9% milk fat, it was not surprising that little or no caesium activity could be detected. Previous studies have indicated that essentially no radioactivity is associated directly with the fat [14, 19, 20, 30, 31]. Our

results confirmed this with <1.2% of the activity from the whole milk transferred to the butteroil.

Powdered samples of whole milk, skim milk and whey were analysed and it was found that the activities of caesium-137 present, exceeded that of the whole milk by factors of 7, 13 and 12, respectively. These are as expected - caesium is not lost in the course of the drying process. An examination of the results in Tb. 4.1.26 confirms this to be the case. With regard to the drying of whey powder, there might at first glance appear to be some loss of caesium. However, the errors quoted are one standard deviation and the 100% transfer falls within this range. The apparent/real loss of caesium may be explained by some powder remaining on the walls of the drying tower and some being lost to the atmosphere during the drying process. Concentrations in whole milk powder lay between 1.8 Bq/kg and 3 Bq/kg, almost two orders of magnitude lower than the levels detected in some milk powders produced within the 2 - 3 week period following the 3rd May 1986, when caesium-137 concentrations in ground level air peaked over Ireland [37].

The remaining samples analysed were all whey derivatives. This is a relatively new area in the dairy industry and may explain why few studies concerning the measurement of caesium transfer to whey products have been reported to date [33, 34, 36]. Some practical problems in the area of sample collection were encountered here: certain techniques had not yet been fully optimized by the creameries and they sometimes had difficulty manufacturing a product to required specifications. A 'line' could therefore be discontinued or altered 'en route' to produce a different product. Consequently it was sometimes difficult to obtain samples of particular products. Secondly, in the routine production of some whey products, e.g., lactose and delactosed whey powder, another problem was encountered. The starting point of the pathway is whey, a mixture of both cheese and casein whey, the exact proportions of which could not be ascertained with any accuracy. The whey is collected and stored for several days until a large enough quantity is available to evaporate down and permit lactose crystallization to occur. The process continues over several days making it extremely difficult to collect samples following on directly one from the other, e.g., the sample of delactosed whey powder collected at the finish may not reflect the sample of

whey taken in at the start. In addition, for administrative purposes, many of the whey samples were collected by creamery staff and not by the author herself.

In spite of these difficulties it was, nevertheless, possible to obtain useful information regarding the transfer of caesium to whey products. About 90% of the caesium activity ends up in the whey, of which 94% by weight is water. Since caesium is a constituent of the mineral content of whey, one would expect a reduction in caesium activity following demineralization. This is, in fact, what was observed. The percentage caesium retained in the (90%) demineralized whey lay between 5 and 10%. Assuming 100% transfer of caesium in the drying of demineralized whey, one should expect to see the same percentage transfer of caesium to demineralized whey powder. The percentage transfer from whey to demineralized whey powder lay between 0.5 and 4.5% which is 60% less than the transfer observed from whey to demineralized whey. A loss of this magnitude in the drying process could not be attributed to the possible sources of loss discussed above and can only be explained by errors in sample collection. In conclusion, what is observed is that a substantial reduction of caesium does occur following demineralization and that at most only 10% of the activity is retained.

Lactose, the sugar component of milk, is crystalized out of the whey. The by-product, known as rennet mother liquor or delactosed whey, when dried, has a relatively high specific activity. The activity for this powder ranged from 5.6 - 22.0 Bq/kg compared to a range of 0.23 - 0.86 Bq/l for the corresponding liquid whey samples. This translates into 72% being transferred from whey to the delactosed whey powder, corresponding to 63% from whole milk. Thus, lactose, with its low activity, retains less than 2% of the activity in the whole milk.

Some industries, e.g., the baking industry, use the delactosed whey powder as a protein supplement. However, a lower mineral content is also required. Attempts were made to produce demineralized delactosed whey from the (90%) demineralized whey. Problems were encountered in producing the specified product, so samples from this pathway could not be collected. An alternative product was manufactured. The whey was evaporated and lactose crystallization

took place. The by-product, the rennet mother liquor, was then partially demineralized (50% demineralization), and referred to as partially demineralized delactosed whey (PDDWY). This was then dried to a powder, known as, partially demineralized delactosed whey powder (PDDWP). The percentage activity retained in this powder was found to be between 22 and 50%. For the PDDWY samples a range of between 4.4 and 11% was transferred, again highlighting the discrepancy which appears to occur in the drying of whey powders. For an accurate determination of the transfer values, accurate yield values are also required, i.e., how many litres of X are required to produce 1 l or 1 kg of product Y. In the case of the partially demineralized whey products, the yield was calculated on the basis of the total solids content in each of the different products and is, therefore, a purely theoretical value. Despite, this however, it is interesting to note that for the delactosed whey powder, 72% transfer of caesium from whey was measured, whereas only 33% was observed for the PDDWP.

In Fig. 4.1.8 the specific activities of four different whey powder products collected at various times of the year are shown (Table 4.1.33¹). The pattern of the results is as expected. The delactosed whey powder (DLW) has the highest concentration, whilst the partially demineralized delactosed whey powder (PDDWP) contained half that of the DLW. Both powders have higher specific activities than the whey powder, the DLW by a factor of four and the PDDWP by a factor of two. The activities in the demineralized whey powder (DWP) are considerably lower, being approximately 10% that of the whey powder.

Some whey protein concentrate (WPC) pathways were also measured, although only one WPC powder sample was obtained. Unfortunately insufficient information regarding these samples was available to determine exact percentage transfers. However, it does appear that a relatively small percentage (<10%) of the original caesium ends up in the WPC. This corresponds with the trend for other high protein samples, e.g., cheese and casein, and agrees well with the 6.6% activity transfer quoted in the literature [33, 34].

As discussed in the literature review, Reavey [21, 22] reported that products

low in caesium-137 were also low in stable potassium, with the ratio of potassium-40 to caesium-137 showing little variation. This is to be expected, given that the ratio of potassium-40 to stable potassium is fixed in nature at 0.0117% [41]. A similar trend was observed in this study. A close examination of Tables 4.1.5 - 4.1.25, where potassium-40 concentrations and potassium-40/caesium-137 ratios are given, demonstrates the consistency of this relationship within each pathway. Where an increase in caesium-137 activity occurs in a product, it is accompanied by an increase of the same magnitude in potassium-40 activity. This suggests that potassium-40 is a useful analog for radiocaesium, particularly when the concentrations of the latter are low.

In those samples where only an MDA value is reported for the caesium activity and hence the potassium-40/caesium-137 ratio can not be calculated, it is possible to determine the caesium concentration and the percentage activity transferred by using the average potassium-40/caesium-137 ratio for the other products in the same pathway. This has been done for a number of different products, e.g., butter, cheese, casein and demineralized whey (Tbs. 4.1.29 - 4.1.32), and the results are found to be in excellent agreement both with those obtained for similar samples in this study (Tbs. 4.1.9, 4.1.12, 4.1.14, 4.1.17) and those reported in the literature Tables 4.1.2 & 4.1.28.

Measured potassium-40 activities in whole milk were found to be fairly constant (~40 Bq/l) throughout the study. This results in a decrease in the potassium-40/caesium-137 ratio with increase in radiocaesium activity.

Caesium-137 concentrations in milk products relative to 1 Bq/kg in whole milk are shown in Fig. 4.1.9 (Table 4.1.34). The results are expressed as Bequerels of caesium per unit weight of product and are relative to liquid milk containing one Bequerel of caesium per unit volume. These data demonstrate how the radioactivity content of milk would be represented in typical dairy products and powders derived from a particular batch of milk. Measurements presented in this format are invaluable to administrators forced to handle an emergency such as Chernobyl, in that it facilitates the interdiction of a particular milk product or products when caesium-137 contamination levels in whole milk exceed previously agreed thresholds. In terms of specific activity (Bq/kg),

products found to have incorporated the least caesium-137 include butter, cream, buttermilk, cheddar cheese, lactose and demineralized whey.

To minimize the level of radiocaesium in dairy products manufactured from contaminated milk, production should be directed away from milk powders, owing to the high concentration factors involved. For short-lived nuclides, an obvious option is storage in product form (usually powder) until radioactive decay has rendered it fit for consumption. This is not a practicable solution for long-lived radionuclides such as strontium-90 or caesium-137. Moreover, Giese [36] has shown that a very expensive operation was required to reduce the caesium content in the whey powder and convert it to a usable form. Where possible, production should be directed towards cream, butter, buttermilk and demineralized products. The resulting skimmilk can be used to make cottage cheese, which Wilson *et al.* [33] and Wood *et al.* [34] have shown contains less than 5% of the activity in whole milk.

Reduction of milk intake by the large-scale substitution of other foods would result in a substantial detriment to nutrition. In practice the intake of strontium-90 cannot be reduced by elimination of milk and milk products from the diet because other foods have, in general, a higher strontium-90 to stable calcium ratio than milk.

From a practical standpoint, it is of interest to consider whether there is any benefit to be gained by the consumption of foods manufactured from contaminated whole milk as compared to consumption of the whole milk itself. For this purpose, the relative contribution of radiostrontium and radiocaesium to the diet by cheddar and cottage cheese has been calculated using the data of Tb. 4.1.35 and tables of food composition provided by Hawk [42]. The radioactivity contribution of cheese to the diet is compared to that of contaminated fresh whole milk when the cheese is supplied in quantities that provide the necessary amounts of either calcium, protein, or calories. The calculations (Tb. 4.1.36) show that when the substitution is made on the basis of the calcium contribution to the diet, the provision of cheddar or cottage cheese in place of the parent fresh whole milk would not alter the amount of radiostrontium ultimately incorporated. The calculations assumed a calcium concentration of 0.87% and 0.05% for the cheddar and

cottage cheese, respectively. The unchanged strontium contribution is to be expected on the basis of the chemical similarity of calcium and strontium, especially during the cheese making process [8]. The radiocaesium intake, however, would be markedly reduced by the use of the substituted products.

When the substitution is based on either an equivalent protein or calorie supply, again no reduction in the radiostrontium intake would be effected by the use of cheddar cheese; cottage cheese, on the other hand, would produce a marked lowering of the radiostrontium intake. The radiocaesium intake would be greatly reduced using either cheddar or cottage cheese to substitute for the whole milk on the basis of equivalent protein or calorie supply (9, 43).

These examples demonstrate that simply changing the form in which milk is incorporated into the diet would not automatically lead to a reduction in the intake of radiocontaminants by the human population. The manufacturing process can produce marked changes, as is shown by the values for radiostrontium and radiocaesium in cottage cheese. This suggests that manufacturing techniques could be worked out that would make it possible, if necessary, to utilize most of the food potential of contaminated milk in a form significantly lower in radionuclide content than the parent whole milk.

Reavey [21, 22] has pointed out that the substitution of cottage cheese for whole milk in the diet would render it deficient in calcium, whilst Garner [44] has asserted that the effect of substituting other foods for whole milk must be considered in relation to their ability to act as alternative sources of calcium, particularly where young children are concerned. For the same reason, butter is no substitute for whole milk in the human diet, even though the percentage transfer of radiocaesium to butter does not exceed 1% .

The distribution of caesium-137 in the various products appears to follow closely that of strontium-90. As was the case with the strontium-90 to stable calcium ratio, the ratio of caesium to stable potassium showed very little variation within the various products. Because of the need for calcium and the greater radiotoxicity of strontium-90, it would appear that if, in times of emergency, the radiocaesium contamination were accompanied by significant radiostrontium (as was the case for weapons fallout), increased hard cheese

production and substitution would not effect a reduction in radiostrontium intake, and any decision as to the utilization of milk or milk products would be based on calcium requirements in the diet. Furthermore, in an emergency it would also be advantageous to expand the production of demineralized whey powders given the proven efficiency of ion-exchange techniques in the removal of radiocaesium and other radioelements from separated whey.

Attempts to remove radionuclides directly from whole milk by the use of ion-exchange resins have met with considerable success [48-60], the main problem to overcome being how to treat the milk without affecting its palatability and nutritional value. An ion-exchange method was developed by analogy with the removal of calcium and strontium from blood to skeleton in the human without altering the net calcium concentration in the former [45, 46, 47]. Strontium was removed from milk by means of a cation exchange resin in a calcium form. The process involved contaminated milk passing through a bed of synthetic resin charged with a mixed solution of metallic ions (calcium, potassium, sodium and magnesium) which are present in milk. An important feature of the process is that the acidity of the milk is adjusted, before passing it through the resin column, from its normal pH of 6.6 down to 5.3 or 5.4. At the normal pH of milk, most of the strontium is bound by other milk constituents and is slow to be exchanged. Passing 25 resin bed volumes of milk at normal pH through the column removes an average of less than 50% of the strontium. At low pH, strontium is largely converted to a soluble and more readily exchangeable form [48].

As early as 1954, Nervik *et al.* [49] used a cation exchange resin in a sodium form to remove both calcium and strontium from milk. This method necessitates the replacement of the calcium content. In 1959, Glueckauf *et al.* [50], employed an anion exchange resin in the chloride form to remove iodide, and suggested that radiostrontium could be removed by passing the milk through a cation exchanger bed, which could be regenerated with a mixture of calcium and sodium chloride. Migicovsky [46] adopting this approach, investigated the removal of strontium and caesium from milk. He found that the same resin could remove both radionuclides but was more efficient for strontium than for caesium. No change in the calcium, potassium or sodium content of the milk, was detected and 86.0% of the strontium was removed by one treatment with

resin. A panel of experts could not detect any change in flavour of the milk as a result of the resin treatment.

Glascock and Bryant [51] have demonstrated, using a pilot cation exchange plant with a capacity of 2,300 l/d [52], that it is feasible to remove radiocaesium from milk with good efficiency under appropriate conditions. Efficiencies in excess of 90% were achieved provided not more than 15 resin bed volumes were treated. However, the effluent milk had to be neutralized prior to consumption. Glascock and Bryant also found that the processed milk retained its nutritional value, with an appreciable deficiency in only one vitamin, thiamine, whose concentration could be restored by the addition of this vitamin afterwards.

Easterly *et al.* investigated the removal of strontium and calcium using two different methods: reverse-flow ion exchange [53] and batch ion exchange [54]. In the former case they found that the calcium form of each resin was more effective in removal of the nuclides than was the sodium form. Between 70% and 80% of strontium-89 and calcium-45 in the skim milk was removed from the second 100 ml passed through the column. Resin preference of strontium over calcium was also shown. For the latter method, namely, batch ion exchange, a study of dosed milk-resin ratio showed that as the ratio increased, the percentage of the removal of strontium-89 and calcium-45 decreased. The percentage removal ranged from a high of 94 and 93 for strontium-89 and calcium-45, respectively, at a milk/resin ratio of 2, to a low of 21% strontium and 16% calcium at a milk/resin ratio of 200.

Marshall *et al.* [55], using a large scale fixed bed ion-exchange system for removal of strontium and iodine found removal efficiencies averaged 98.9% for iodine-131, 94.6% strontium-85 and 90.0% for strontium-90. They also looked at the removal of caesium and found that the resin removed 66% of caesium-137. Tests conducted on the processed milk indicated some decrease in flavour score, particularly in the first milk passed through the anion resin column. Bacterial studies indicated that a slight increase in psychrophil counts occurred during processing. Heinemann *et al.* [56] also found a slight decrease in flavour score along with minor compositional changes in freezing point, curd tension, titratable acidity and ash. There was an increase of

0.36% in concentration of potassium citrate, due to acidification and neutralization of the milk. Insignificant changes were observed in the other components tested. The same group operated the plant over a ten month period [57] and found that the milk was organoleptically acceptable. The processed milk was spray dried and utilized for animal feed. They concluded that in the event of environmental contamination exceeding safe tolerances, this process is a reasonable and practicable means for substantially reducing the levels of strontium-90 in milk.

Murthy [58] employed standard procedures to make cultured milk, butter and Cheddar cheese from resin-treated milk and reported that (i) with properly selected cultures, cultured milk could be prepared satisfactorily from the treated milk; (ii) butter made from resin-treated milk had better keeping quality at room temperature and (iii) other than a slight improvement in flavour, there were no significant differences in some of the observed physical and chemical properties of control and treated Cheddar cheese samples. Murthy *et al.* [59], in their investigation of the removal of iodine-131 from milk found that approximately $98 \pm 2\%$ was removed from 120-130 resin bed volumes and that all the iodine-131 was removed except the protein-bound iodine-131, which was not available for ion-exchange. Although the protein-bound iodine in milk may vary for individual cows (0 - 10%), it is negligible in market milk contaminated by fresh fallout.

Isaacks *et al.* [60] undertook a chemical analysis of processed milk for nutritional evaluation. A comparison of results obtained on control and resin-treated milk samples showed no significant change in total solids, butterfat, protein, flavour quality and vitamins A and B12. The ash, potassium and citric acid content of the resin-treated milk increased by approximately 14%, 80% and 100%, respectively, whilst the thiamine, niacin and vitamin B6 content of the processed milk decreased by 50%, 27% and 15%, respectively. The data indicate that 86% of the free thiamine and 20% of the bound thiamine are removed from the milk by the resin treatment, in agreement with the results reported by Glascock and Bryant [51].

Edmondson *et al.* [48] described a pilot plant process developed in 1962 for strontium removal. While a great deal of research went into the development

of the plant, improvements were necessary and still are today, to assure nutritionally satisfactory milk, to evaluate possible sanitation problems and to reduce the cost. This last point was also highlighted by Migicovsky [61], who raised the question as to whether the process could be used on a commercial basis in the event of an emergency. Its economic viability remains a point of debate even today, although, under extreme conditions, such a course of action might be essential.

4.1.5. SUMMARY AND CONCLUSIONS

The pasture-cow-milk-man pathway is highly sensitive to air-borne pollution as was exemplified by the Windscale and more recently the Chernobyl accident, when much of the milk produced in Europe was contaminated - mainly by radiocaesium and radiiodine.

Iodine, with its relatively short half-life of 8 days, ceases to be important after 7-8 weeks following a pulsed release. Caesium, on the other hand, with a half-life of 30 years, can be measured in milk and its products for decades following ground contamination.

An extensive literature survey revealed that a considerable amount of work had been conducted concerning the transfer of radionuclides from milk to its products. The principal nuclides investigated were strontium, iodine and caesium, with more emphasis on strontium, as this constituted the greater hazard following the nuclear weapons testing in the late 1950s and early 1960s, which signaled the onset of this line of research.

Studies carried out prior to Chernobyl concentrated on activity transfers from whole milk to skimmilk, cream, butter and cheese, with little else being investigated, as these were the principal commercial products for human consumption. It was demonstrated that caesium and strontium behave similarly, except in the manufacture of cheese, where most of the strontium is transferred to the cheese and only about 10% of the caesium. The studies revealed that caesium travels with the aqueous fractions, e.g., skimmilk (94%), leaving the fat fractions relatively depleted in caesium, e.g., cream (6%) and butter (~2%). Products with a high protein content also receive little of the caesium present in the whole milk. Some researchers also looked at the behaviour of calcium and potassium and found that they behaved similarly to strontium and caesium, respectively.

Post-Chernobyl research confirmed the findings of the 1960s but also highlighted gaps in our knowledge as the dairy industry had developed extensively and many new products were now being produced in commercial quantities. This was the case especially for whey and its products which up

until about 15 years previously had been considered a waste product. In this study emphasis was given to this area of the dairy industry.

The findings of this study are in close accord with previous work (Table 28). These demonstrate that about 90% of the caesium in whole milk ends up in the whey which, when further processed, yields a product, known as delactosed whey powder, whose specific activity is more than 20 times that of the whole milk.

The expansion of the whey industry is by no means complete, with further development of new products and their uses envisaged. Consequently, it will be necessary to complement the existing database as these products reach the market place.

With regard to the more traditional milk products, the picture is clear. The drying of powders merely serves to increase the radionuclide concentrations and is of little value unless the contaminants have short half-lives. In this case, the storing of powders until the activities have decayed to the permissible levels decided upon by the relevant authorities following an accidental radioactivity release, would be a useful course of action. If the contaminants have long half-lives, e.g., caesium or strontium, different counter-measures will be necessary the nature of which will be determined by the levels of contamination involved.

Shortly after the Chernobyl accident, the Community was faced with the problem of contaminated foodstuffs. In May 1986, following the accident, the Council of the European Communities adopted Regulation (EEC) No. 1707/86 [62] governing imports of agricultural products originating in third countries following the accident at the Chernobyl nuclear power station. This Regulation set maximum levels of 370 Bq/kg, total caesium, for milk and foodstuffs intended for infants under 6 months, and 600 Bq/kg for all other products. The Regulation was extended on 30 September 1986 by Regulation (EEC) No. 3020/86 [63] and further extended by Regulation (EEC) No. 624/87 [64] until 31 October 1987.

On 22 December 1987 two Regulations were adopted by the Council: Regulation

(Euratom) No. 3954/87 [65] and Regulation (EEC) No. 3955/87 [66]. The former, is a regulation to be invoked in the event of a future accident or radioactive contamination of foodstuffs. It lays down maximum permitted levels of radioactive contamination of foodstuffs and of feedingstuffs following a nuclear accident or any other kind of radiological emergency. The latter, is a further extension until December 1989 of the original Regulation (EEC) No. 1707/86, and states that "without prejudice to Council Regulation (Euratom) No. 3954/87 of 22 December 1987, the Community must continue to ensure, with regard to the specific effects of the accident at Chernobyl, that agricultural products and processed agricultural products intended for human consumption and likely to be contaminated are introduced into the Community only according to common arrangements".

In July 1988, by Council Regulation (EEC) No. 1983/88 [67], rules were laid down for the application of Regulation (EEC) No. 3955/87. In December 1989 this Regulation was further extended to 31 March 1990 by Regulation (EEC) No.4003/89 [68]. On 22 March 1990 Council Regulation (EEC) No.737/90 [69] was adopted which extends the 370 Bq/kg and 600 Bq/kg, total caesium limits, for milk and baby foods, and other foodstuffs, respectively, until 31 March 1995. This Regulation was adopted without prejudice to the possibility of resorting where necessary, in the future, to the provisions of Council Regulation (Euratom) No. 3954/87 of 22 December 1987 [65] as amended by Regulation (Euratom) No. 2218/89 of 18 July 1989 [70] and further amended by Regulation (Euratom) No. 770/90 of 29 March 1990 [71]. The maximum permitted levels for babyfoods, dairy produce, other foods except minor foodstuffs, liquid foodstuffs and feedingstuffs as laid down by this Regulation, for future emergencies, are given in Table 4.1.37.

The maximum permitted levels for dairy products, as laid down in Regulation No.3954/87, are calculated on the basis of the reconstituted product as ready for consumption. This facilitates a decision as to which lines of production can be pursued following contamination of milk. It is important also to bear in mind which radionuclides are present owing to their different behavioural patterns during milk processing and the different hazards associated with each. Account must also be taken of dietary requirements when replacing one dairy product with another.

If the level of contamination in milk exceeds the permissible limits, direct decontamination may be necessary. Before this latter option is adopted, an evaluation of alternative processes would have to be carried out to justify the expenditure. However, the production of demineralized whey products could be expanded as these facilities are available in many creameries.

It is not possible to state categorically what procedures ought to be adhered to following the contamination of milk, as each situation is likely to be different with many factors coming into play. What this study has served to do, however, is to demonstrate how caesium behaves in the course of processing and, in particular, to provide realistic transfer factors from milk to a wide range of milk products. The latter enable the relevant authorities to make decisions as how best to optimize the use of milk in the event of it becoming contaminated.

A final point worth mentioning, is that the levels incorporated in any Community system must have public and political credibility bearing in mind the very high level of public concern which is likely to prevail. One major aspect of this credibility, is the relationship of the levels set by the Community to those in use elsewhere in the world. These should be harmonised to the greatest degree possible.

In conclusion, this and previous studies have shown that caesium concentrates in the aqueous fractions of milk leaving the fat fractions relatively depleted of radioactivity. In the event of milk becoming contaminated, production should be diverted away from powders. Production should concentrate on the demineralized whey products as the caesium is removed along with other minerals in the demineralization process. If the milk is heavily contaminated and if the accident is of such a widespread nature that alternative dietary substitutes are also heavily contaminated, consideration should be given to direct decontamination of the milk. When replacing one milk product in the diet with another, it is necessary to ensure that the nutritional dietary requirements are met.

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TABLE 4.1.1
Quantitative Composition of Milk (1)

Constituent	Limits of Variation %	Mean Value %
Water	85.5 - 89.5	87.5
Total solids	10.5 - 14.5	13.0
Fat	2.5 - 6.0	3.9
Proteins	2.9 - 5.0	3.4
Lactose	3.6 - 5.5	4.8
Minerals	0.6 - 0.9	0.8

TABLE 4.1.2
Summary from Literature Review of the Percentage Transfer of Radiocaesium to a Range of Dairy Products

Product	Antila <i>et al</i> [29]	Assimakopoulos <i>et al</i> [10,11]	Calapaj & Ongaro [15]	Everitt & Paulsson [28]	Kandarakis & Anifantakis [27]	Kankare <i>et al</i> [30,31]	Kirchmann <i>et al</i> [16]	Lagoni <i>et al</i> [19,20]	Lengemann [9]	Ohmomo & Tsugo [14]	Pirhonen <i>et al</i> [32]	Reavey [21,22]	Wilson <i>et al</i> [33]	Range
Whole Milk	100	100	100	100	100	100	100	100	100	100	100	100	100	
Cream					3	5.9		16	6	7		6	2.7	3-16
Skim milk	91					91		85	91	92		91	99	85-99
Butter	0.7		<1.5			0.8		2.2	0.3			<1	0.4	0.3-2.2
Buttermilk	5.3					5.1		13					2.3	2-13
Butterfat						0		0		0				0
Cheese (Rennet)	11	7.4 ^a	<10	5	9 ^b		5-23 ^c		7.7	9.6	6		4.5	1-23
Cheese Whey	89	71 ^b					77-96 ^c			89	93		93	71-96
Sour Cheese	12						12							12
Sour Cheese Whey	88						88							88
Cottage Cheese					4 ^d				1.1	4.7		1		1-5
Cottage Cheese Whey										70		82	95	70-95
Acid Casein						4.0	1	1.6						1-4
Acid Casein Whey						90	84	83						83-90
Whey Proteins						1.0		1.4						1.0-1.4
Deproteinized Acid Whey							89		82					82-89
Rennet Casein						5.7	7.7	1.8						1.8-7.7
Rennet Casein Whey						88	77	83						77-88
Rennet Whey Proteins						0.6		2.4						0.6-2.4
Deproteinized Rennet Whey							88	80						80-88
Demineralized Whey													<1.5	<1.5
Whey Protein Concentrate													6.6	6.6
Permeate													8.8	8.8
Yoghurt (Skim milk)					34									34

a) Gruyere Cheese (Fwe's milk)
b) Whey after removal of cream
c) Kefalogryni Cheese
d) Myzithra (Whey Cheese)
e) Different values for different types of cheese

TABLE 4.1.3
Dairy Products as Food Ingredients

Product	Bulk Butter	Butter Oil	Bulk Cheese	Whole Milk Powder	Skim Milk Powder	Fat Filled Milk	Whey Powder	Butter Milk Powder	Caseinate	Chocolate Crumb	Frozen Cream
Baby Foods	✓	✓		✓	✓	✓	✓	✓	✓		
Bakery Goods	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓
Beverages	✓	✓		✓	✓	✓	✓	✓	✓	✓	
Confectionary	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓
Desserts	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓
Dietetic Foods			✓	✓	✓	✓	✓	✓	✓		
Dry Mixes	✓	✓	✓	✓	✓	✓	✓	✓	✓		
Ice Cream	✓	✓		✓	✓	✓	✓	✓		✓	✓
Processed Meats	✓	✓	✓		✓	✓	✓	✓	✓		
Soups/Sauces	✓	✓	✓	✓	✓	✓	✓	✓	✓		✓
Pharmaceutical					✓	✓	✓		✓		

Source: An Bord Bainne Diary, Bord Bainne, Dublin, 1988

TABLE 4.1.4
Number of Samples Collected at each Creamery*

Creamery	A		B		C	
Month	1988	1989	1988	1989	1988	1989
February	—	16	—	—	—	9
March	30	—	38	27	—	—
April	38	16	—	—	—	—
May	—	—	—	—	—	12
June	14	16	23	—	11	—
July	—	—	—	20	—	12
August	12	19	—	—	—	—
September	—	—	—	—	—	—
October	31	16	22	—	14	3

*A, B and C are code letters for the three creameries used in the present study

TABLE 4.1.5

Cs-137 and K-40 Concentrations in Whole Milk and Skimmilk and the Percentage Transfer of Cs-137 from Whole Milk to Skimmilk

Product	Cs-137 (Bq/l)	% Cs-137 transfer	K-40 (Bq/l)	K-40/Cs-137
Whole Milk	1.1 ± 0.2		39 ± 2	35 ± 5
Skimmilk	1.1 ± 0.2	91 ± 18	33 ± 2	30 ± 5
Whole Milk	0.34 ± 0.07		43 ± 1	127 ± 25
Skimmilk	0.30 ± 0.05	79 ± 21	47 ± 1	160 ± 27
Whole Milk	0.21 ± 0.05		18 ± 1	85 ± 20
Skimmilk	0.32 ± 0.04	139 ± 38	49 ± 1	152 ± 21
Whole Milk	0.23 ± 0.05		40 ± 1	172 ± 38
Skimmilk	0.23 ± 0.05	89 ± 27	34 ± 1	147 ± 31
Whole Milk	0.25 ± 0.13		43 ± 1	172 ± 91
Skimmilk	0.21 ± 0.05	76 ± 44	42 ± 1	198 ± 46
Whole Milk	0.23 ± 0.08		45 ± 1	195 ± 68
Skimmilk	0.24 ± 0.02	95 ± 34	42 ± 1	197 ± 18
Whole Milk	0.28 ± 0.12		50 ± 3	179 ± 77
Skimmilk	0.24 ± 0.06	78 ± 39	46 ± 1	190 ± 49

TABLE 4.1.6

Cs-137 and K-40 Concentrations in Whole Milk and Cream and the Percentage Transfer of Cs-137 from Whole Milk to Cream

Product	Cs-137 (Bq/l)	% Cs-137 transfer	K-40 (Bq/l)	K-40/Cs-137
Whole Milk	1.1 ± 0.2		39 ± 2	35 ± 5
Cream	0.64 ± 0.18	5 ± 2	14 ± 1	22 ± 6
Whole Milk	0.34 ± 0.07		43 ± 1	127 ± 25
Cream	0.18 ± 0.05	5 ± 2	23 ± 1	128 ± 37
Whole Milk	0.21 ± 0.05		18 ± 1	85 ± 20
Cream	0.26 ± 0.04	11 ± 3	26 ± 1	101 ± 17
Whole Milk	0.23 ± 0.05		40 ± 1	172 ± 38
Cream	0.15 ± 0.09	6 ± 4	19 ± 1	133 ± 78
Whole Milk	0.25 ± 0.13		43 ± 1	172 ± 91
Cream	0.16 ± 0.04	6 ± 4	22 ± 1	137 ± 37
Whole Milk	0.23 ± 0.08		45 ± 1	195 ± 68
Cream	0.20 ± 0.02	8 ± 3	27 ± 2	133 ± 16
Whole Milk	0.28 ± 0.12		50 ± 3	179 ± 77
Cream	0.16 ± 0.04	5 ± 3	21 ± 1	130 ± 33

TABLE 4.1.7

Cs-137 and K-40 concentrations in Whole Milk and Whole Milk Powder and the Percentage Transfer of Cs-137 from Whole Milk to Whole Milk Powder

Product	Cs-137 (Bq/kg) or (Bq/l)	% Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40/Cs-137
Whole Milk	0.30 ± 0.15		46 ± 2	154 ± 78
Whole Milk Powder	2.3 ± 0.5	97 ± 52	359 ± 4	157 ± 31
Whole Milk	0.24 ± 0.07		47 ± 2	194 ± 60
Whole Milk Powder	1.9 ± 0.2	99 ± 33	373 ± 4	197 ± 20
Whole Milk	0.24 ± 0.03		52 ± 2	218 ± 28
Whole Milk Powder	1.8 ± 0.3	96 ± 20	370 ± 4	205 ± 33
Whole Milk	0.44 ± 0.10		50 ± 6	115 ± 25
Whole Milk Powder	3.1 ± 0.3	87 ± 21	382 ± 7	125 ± 13
Whole Milk	0.39 ± 0.22		42 ± 2	107 ± 60
Whole Milk Powder	2.2 ± 0.2	71 ± 41	310 ± 6	140 ± 11
Whole Milk	0.28 ± 0.09		55 ± 3	195 ± 60
Whole Milk Powder	2.1 ± 0.2	95 ± 29	301 ± 6	143 ± 11
Whole Milk	0.54 ± 0.06		68 ± 3	126 ± 16
Whole Milk Powder	3.0 ± 0.4	69 ± 12	374 ± 4	127 ± 15

TABLE 4.1.8

Cs-137 and K-40 Concentrations in Skimmilk and Skimmilk Powder and the Percentage Transfer of Cs-137 from Skimmilk to Skimmilk Powder

Product	Cs-137 (Bq/kg) or (Bq/l)	% Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40/Cs-137
Skimmilk	0.38 ± 0.19		24 ± 1	62 ± 30
Skimmilk Powder	4.4 ± 0.4	104 ± 52	508 ± 5	116 ± 9
Skimmilk	0.30 ± 0.05		47 ± 1	159 ± 27
Skimmilk Powder	3.6 ± 0.3	110 ± 21	485 ± 5	135 ± 11
Skimmilk	0.32 ± 0.04		49 ± 1	152 ± 21
Skimmilk Powder	5.2 ± 0.4	147 ± 24	498 ± 5	96 ± 7

TABLE 4.1.9

Cs-137 and K-40 Concentrations in Cream and Butter and the Percentage Transfer of Cs-137 from Cream to Butter

Product	Cs-137 (Bq/kg) or (Bq/l)	% Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40/Cs-137
Cream	0.46 ± 0.05		18 ± 1	40 ± 5
Butter	<0.3	<29	6.6 ± 0.8	>24
Cream	0.28 ± 0.04		24 ± 1	84 ± 14
Butter	<0.4	<61	7.7 ± 1.2	>22
Cream	0.29 ± 0.06		23 ± 1	79 ± 17
Butter	<0.4	<58	13 ± 1	>36
Cream	0.15 ± 0.04		24 ± 2	156 ± 39
Butter	<0.2	<53	5.8 ± 0.6	>93
Cream	0.23 ± 0.05		21 ± 2	92 ± 18
Butter	<0.4	<83	<2	>4

TABLE 4.1.10

Cs-137 and K-40 Concentrations in Cream and Buttermilk and the Percentage Transfer of Cs-137 from Cream to Buttermilk

Product	Cs-137 (Bq/l)	% Cs-137 transfer	K-40 (Bq/l)	K-40/Cs-137
Cream	0.46 ± 0.05		18 ± 1	40 ± 5
Buttermilk	0.47 ± 0.11	51 ± 13	31 ± 2	67 ± 17
Cream	0.28 ± 0.04		24 ± 1	84 ± 14
Buttermilk	0.47 ± 0.05	84 ± 16	30 ± 2	64 ± 7
Cream	0.29 ± 0.06		23 ± 1	79 ± 17
Buttermilk	0.25 ± 0.05	44 ± 12	39 ± 1	154 ± 28
Cream	0.15 ± 0.04		24 ± 1	156 ± 39
Buttermilk	0.26 ± 0.05	85 ± 28	44 ± 1	168 ± 35

TABLE 4.1.11

Cs-137 and K-40 Concentrations in Cream and Butteroil and the Percentage Transfer of Cs-137 from Cream to Butteroil

Product	Cs-137 (Bq/l)	% Cs-137 transfer	K-40 (Bq/l)	K-40/Cs-137
Cream Butteroil	0.08 ± 0.03 <0.03	<18	12 ± 1 <2	144 ± 55 >75
Cream Butteroil	0.15 ± 0.09 <0.04	<13	19 ± 1 <2	133 ± 78 >43
Cream Butteroil	0.16 ± 0.04 <0.04	<12	21 ± 1 <2	130 ± 33 >44

TABLE 4.1.12

Cs-137 and K-40 Concentrations in Skimmilk and Casein and the Percentage Transfer of Cs-137 from Skimmilk to Casein

Product	Cs-137 (Bq/kg) or (Bq/l)	% Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40/Cs-137
Skimmilk Casein	1.0 ± 0.1 <0.2	<0.5	34 ± 2 2.4 ± 0.6	34 ± 3 >14
Skimmilk Casein	0.55 ± 0.23 <1	<0.7	68 ± 1 13 ± 1	124 ± 51 >101
Skimmilk Casein	0.26 ± 0.03 <0.2	<2	43 ± 1 5.4 ± 0.5	165 ± 21 >30
Skimmilk Casein	0.24 ± 0.02 <0.2	<3	47 ± 1 4.8 ± 1.5	197 ± 18 >23
Skimmilk Casein	0.24 ± 0.06 <0.1	<1	46 ± 1 4.3 ± 0.6	190 ± 49 >43

TABLE 4.1.13

Cs-137 and K-40 Concentrations in Skimmilk and Casein Whey and the Percentage Transfer of Cs-137 from Skimmilk to Casein Whey

Product	Cs-137 (Bq/l)	% Cs-137 transfer	K-40 (Bq/l)	K-40/Cs-137
Skimmilk	1.0 ± 0.1		34 ± 2	34 ± 3
Casein Whey	0.69 ± 0.06	65 ± 8	41 ± 2	59 ± 6
Skimmilk	0.55 ± 0.23		68 ± 1	124 ± 51
Casein Whey	0.39 ± 0.90	67 ± 31	47 ± 1	120 ± 28
Skimmilk	0.26 ± 0.03		43 ± 1	165 ± 20
Casein Whey	0.27 ± 0.05	98 ± 22	42 ± 1	155 ± 28
Skimmilk	0.24 ± 0.02		47 ± 1	197 ± 18
Casein Whey	0.28 ± 0.08	110 ± 32	49 ± 1	175 ± 49
Skimmilk	0.24 ± 0.06		46 ± 1	190 ± 49
Casein Whey	0.28 ± 0.05	110 ± 35	43 ± 1	154 ± 29

TABLE 4.1.14

Cs-137 and K-40 Concentrations in Whole Milk and Cheese and the Percentage Transfer of Cs-137 from Whole Milk to Cheese

Product	Cs-137 (Bq/kg) or (Bq/l)	% Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40/Cs-137
Whole Milk	0.63 ± 0.09		40 ± 3	63 ± 10
Cheese	<0.3	<5	30 ± 2	>99
Whole Milk	0.35 ± 0.07		40 ± 2	114 ± 22
Cheese	0.38 ± 0.12	11 ± 4	22 ± 1	57 ± 18
	0.37 ± 0.03	10 ± 2	23 ± 1	62 ± 6
Whole Milk	0.29 ± 0.02		43 ± 2	148 ± 13
Cheese	<0.3	<3	20 ± 2	>61
Whole Milk	0.26 ± 0.07		46 ± 1	176 ± 46
Cheese	<0.3	<3	23 ± 1	>82
Whole Milk	0.18 ± 0.03		44 ± 1	246 ± 34
Cheese	0.30 ± 0.07	5.0 ± 1.4	44 ± 1	145 ± 33
Whole Milk	0.18 ± 0.06		41 ± 1	225 ± 70
Cheese	0.31 ± 0.07	5.1 ± 1.9	23 ± 3	74 ± 16
Whole Milk	0.25 ± 0.07		45 ± 1	179 ± 50
Cheese	<0.3	<4	16 ± 2	>52
Whole Milk	0.18 ± 0.06		41 ± 1	221 ± 75
Cheese	0.17 ± 0.07	2.7 ± 1.5	19 ± 1	116 ± 51

TABLE 4.1.15

Cs-137 and K-40 Concentrations in Whole Milk and Cheese Whey and the Percentage Transfer of Cs-137 from Whole Milk to Cheese Whey

Product	Cs-137 (Bq/l)	% Cs-137 transfer	K-40 (Bq/l)	K-40/Cs-137
Whole Milk	0.63 ± 0.09		40 ± 3	63 ± 10
Cheese Whey	0.60 ± 0.02	92 ± 13	41 ± 2	68 ± 3
Whole Milk	0.35 ± 0.07		40 ± 2	114 ± 22
Cheese Whey	0.38 ± 0.09	103 ± 31	40 ± 2	106 ± 27
Whole Milk	0.29 ± 0.02		43 ± 2	148 ± 13
Cheese Whey	0.26 ± 0.09	85 ± 48	43 ± 1	165 ± 83
Whole Milk	0.26 ± 0.07		46 ± 1	176 ± 46
Cheese Whey	0.25 ± 0.05	91 ± 29	45 ± 1	181 ± 33
Whole Milk	0.18 ± 0.03		44 ± 1	246 ± 34
Cheese Whey	0.19 ± 0.04	100 ± 25	44 ± 1	231 ± 49
Whole Milk	0.18 ± 0.06		41 ± 1	225 ± 70
Cheese Whey	0.16 ± 0.04	84 ± 34	38 ± 2	235 ± 59
Whole Milk	0.25 ± 0.07		45 ± 1	179 ± 50
Cheese Whey	0.16 ± 0.06	60 ± 28	32 ± 1	200 ± 76
Whole Milk	0.18 ± 0.06		41 ± 1	221 ± 75
Cheese Whey	0.21 ± 0.07	109 ± 51	40 ± 1	325 ± 107

TABLE 4.1.16
Cs-137 and K-40 Concentrations in Whey and Whey Powder and the Percentage Transfer of Cs-137 from Whey to Whey Powder

Product	Cs-137 (Bq/kg) or (Bq/l)	% Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40/Cs-137
Whey	0.54 ± 0.06		36 ± 1	66 ± 8
Whey Powder	7.2 ± 0.4	74 ± 10	653 ± 7	91 ± 5
Whey	0.32 ± 0.10		47 ± 1	146 ± 44
Whey Powder	6.9 ± 1.0	119 ± 40	994 ± 20	145 ± 22
Whey (concentrate)	3.9 ± 1.0		504 ± 10	127 ± 29
Whey Powder	5.8 ± 0.5	82 ± 20	679 ± 7	117 ± 11
Whey	0.29 ± 0.16		32 ± 2	111 ± 62
Whey Powder	3.5 ± 0.2	66 ± 37	617 ± 6	176 ± 12
Whey	0.28 ± 0.05		43 ± 1	154 ± 29
Whey Powder	3.3 ± 1.4	66 ± 30	647 ± 13	196 ± 80
Whey	0.25 ± 0.01		45 ± 1	178 ± 9
Whey Powder	2.1 ± 0.1	47 ± 4	562 ± 11	265 ± 16
Whey	0.41 ± 0.06		44 ± 1	107 ± 17
Whey Powder	6.6 ± 0.7	90 ± 17	681 ± 14	104 ± 11

TABLE 4.1.17
Cs-137 and K-40 Concentrations in Whey and Demineralized Whey and the Percentage Transfer of Cs-137 from Whey to Demineralized Whey

Product	Cs-137 (Bq/l)	% Cs-137 transfer	K-40 (Bq/l)	K-40/Cs-137
Whey	0.37 ± 0.06		46 ± 1	123 ± 18
Demineralized Whey	<0.06	<16	2.4 ± 0.2	>40
Whey	0.29 ± 0.16		32 ± 2	154 ± 86
Demineralized Whey	<0.04	<12	0.33 ± 0.03	>9
Whey	0.40 ± 0.03		54 ± 1	137 ± 11
Demineralized Whey	0.02 ± 0.01	5 ± 3	2.8 ± 1.7	138 ± 68
Whey	0.20 ± 0.01		53 ± 1	264 ± 13
Demineralized Whey	0.02 ± 0.01	9 ± 3	2.3 ± 1.1	127 ± 39
Whey	0.25 ± 0.14		45 ± 1	178 ± 9
Demineralized Whey	0.02 ± 0.01	91 ± 8	3.5 ± 0.2	161 ± 31

TABLE 4.1.18

Cs-137 and K-40 Concentrations in Whey and Demineralized Whey Powder and the Percentage Transfer of Cs-137 from Whey to Demineralized Whey Powder

Product	Cs-137 (Bq/kg) or (Bq/l)	% Cs-137 transfer	K-40 (Bq/kg) or (Bq l)	K-40 Cs-137
Whey	0.29 ± 0.16		32 ± 2	154 ± 86
Demin. Whey Powder	<0.2	<2	22 ± 1	>177
Whey	0.25 ± 0.01		45 ± 1	178 ± 9
Demin. Whey Powder	0.26 ± 0.02	4.5 ± 0.4	101 ± 2	391 ± 27
Whey	0.28 ± 0.05		43 ± 1	154 ± 29
Demin. Whey Powder	0.22 ± 0.09	3.5 ± 1.6	33 ± 2	151 ± 63
Whey	0.40 ± 0.03		54 ± 1	137 ± 11
Demin. Whey Powder	0.07 ± 0.02	0.80 ± 0.18	33 ± 1	477 ± 105
Whey	0.20 ± 0.01		53 ± 1	264 ± 13
Demin. Whey Powder	0.11 ± 0.03	2.4 ± 0.6	28 ± 1	255 ± 51

TABLE 4.1.19

Cs-137 and K-40 Concentrations in Whey and Rennet Mother Liquor (RML) and the Percentage Transfer of Cs-137 from Whey to RML

Product	Cs-137 (Bq/l)	% Cs-137 transfer	K-40 (Bq/l)	K-40/Cs-137
Whey	0.86 ± 0.15		46 ± 2	53 ± 10
RML	8.3 ± 0.3	56 ± 10	501 ± 25	61 ± 4
Whey	0.52 ± 0.25		35 ± 2	67 ± 33
RML	4.4 ± 0.2	50 ± 24	41 ± 2	93 ± 6
Whey	0.34 ± 0.10		45 ± 1	132 ± 40
RML	3.3 ± 0.2	57 ± 18	465 ± 9	141 ± 8
Whey	0.57 ± 0.07		45 ± 1	79 ± 9
RML	3.2 ± 0.5	32 ± 6	494 ± 15	156 ± 23
Whey	0.23 ± 0.09		33 ± 1	141 ± 55
RML	2.3 ± 0.2	60 ± 2	355 ± 7	152 ± 12
Whey	0.40 ± 0.03		54 ± 1	137 ± 11
RML	4.7 ± 0.1	119 ± 10	689 ± 14	145 ± 4

TABLE 4.1.20

Cs-137 and K-40 Concentrations in Whey and Lactose and the Percentage Transfer
of Cs-137 from Whey to Lactose

Product	Cs-137 (Bq/kg) or (Bq/l)	% Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40 Cs-137
Whey Lactose	0.86 ± 0.15 0.58 ± 0.09	2.0 ± 0.5	46 ± 2 21 ± 1	53 ± 10 35 ± 6
Whey Lactose	0.52 ± 0.25 0.23 ± 0.06	1.3 ± 0.7	35 ± 2 17 ± 1	67 ± 33 74 ± 19
Whey Lactose	0.34 ± 0.10 <0.2	<1	45 ± 1 16 ± 1	132 ± 40 >107
Whey Lactose	0.57 ± 0.07 <0.1	<6	45 ± 1 12 ± 1	79 ± 10 >97
Whey Lactose	0.23 ± 0.09 <0.1	<1	33 ± 1 18 ± 1	141 ± 55 >220
Whey Lactose	0.28 ± 0.08 0.29 ± 0.06	2.9 ± 1.0	49 ± 1 15 ± 1	175 ± 49 51 ± 12
Whey Lactose	0.24 ± 0.01 0.05 ± 0.01	0.60 ± 0.13	50 ± 1 15 ± 1	211 ± 13 321 ± 67
Whey Lactose	0.40 ± 0.03 0.09 ± 0.02	0.66 ± 0.17	54 ± 1 10 ± 1	137 ± 11 106 ± 28
Whey Lactose	0.25 ± 0.01 0.04 ± 0.01	0.40 ± 0.12	45 ± 1 10 ± 1	178 ± 9 250 ± 75

TABLE 4.1.21

Cs-137 and K-40 Concentrations in Whey and Delactosed Whey Powder (DLW) and the Percentage Transfer of Cs-137 from Whey to DLW

Product	Cs-137 (Bq/l)	% Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40/Cs-137
Whey	0.86 ± 0.15		46 ± 2	53 ± 10
DLW	22 ± 1	73 ± 13	1367 ± 27	62 ± 2
Whey	0.23 ± 0.09		33 ± 1	141 ± 55
DLW	6.7 ± 1.1	84 ± 36	1240 ± 12	184 ± 31
Whey	0.28 ± 0.08		49 ± 1	175 ± 49
DLW	7.1 ± 1.3	73 ± 24	1308 ± 13	184 ± 33
Whey	0.24 ± 0.01		50 ± 1	211 ± 13
DLW	5.6 ± 0.2	68 ± 5	1220 ± 24	217 ± 9
Whey	0.40 ± 0.03		54 ± 1	137 ± 11
DLW	7.2 ± 0.4	52 ± 5	1069 ± 21	149 ± 9
Whey (conc)	2.8 ± 0.1		458 ± 9	161 ± 8
DLW	8.1 ± 0.5	82 ± 6	1391 ± 28	171 ± 10

TABLE 4.1.22

Cs-137 and K-40 Concentrations in Whey and Partially Demineralized Delactosed Whey (PDDWY) and the Percentage Transfer of Cs-137 from Whey to PDDWY

Product	Cs-137 (Bq/l)	% Cs-137 transfer	K-40 (Bq/l)	K-40/Cs-137
Whey	0.25 ± 0.01		45 ± 1	178 ± 9
PDDWY	0.48 ± 0.02	11 ± 1	90 ± 2	188 ± 8
Whey	0.40 ± 0.03		55 ± 1	137 ± 11
PDDWY	0.30 ± 0.02	4.4 ± 0.5	45 ± 1	151 ± 12
Whey	0.21 ± 0.02		45 ± 1	210 ± 21
PDDWY	0.31 ± 0.02	8.6 ± 1.0	72 ± 1	231 ± 16

TABLE 4.1.23

Cs-137 and K-40 Concentrations in Whey and Partially Demineralized Delactosed Whey Powder (PDDWP) and the Percentage Transfer of Cs-137 from Whey to PDDWP

Product	Cs-137 (Bq/kg) or (Bq/l)	% Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40/Cs-137
Whey	0.25 ± 0.03		45 ± 1	178 ± 9
PDDWP	4.4 ± 0.3	50 ± 4	765 ± 8	173 ± 12
Whey	0.40 ± 0.03		54 ± 8	137 ± 11
PDDWP	3.1 ± 0.1	22 ± 2	451 ± 9	145 ± 6
Whey	0.21 ± 0.02		45 ± 1	210 ± 21
PDDWP	2.1 ± 0.1	28 ± 3	485 ± 10	232 ± 9

TABLE 4.1.24

Cs-137 and K-40 Concentrations in Whey and Whey Protein Concentrate (WPC)

Product	Cs-137 (Bq/kg) or (Bq/l)	K-40 (Bq/kg) or (Bq/l)	K-40/Cs-137
Whey	0.29 ± 0.16	32 ± 1	111 ± 62
WPC	0.35 ± 0.05	39 ± 1	118 ± 15
Whey	0.20 ± 0.01	53 ± 1	264 ± 16
WPC	0.97 ± 0.31	239 ± 5	246 ± 10
Whey	0.30 ± 0.02	51 ± 1	171 ± 12
WPC	0.09 ± 0.01	25 ± 1	263 ± 26

TABLE 4.1.25
Cs-137 and K-40 Concentrations in Whey and Permeate

Product	Cs-137 (Bq/l)	K-40 (Bq/l)	K-40/Cs-137
Whey	0.29 ± 0.16	32 ± 1	111 ± 62
Permeate	1.3 ± 0.1	171 ± 3	136 ± 12
Whey	0.20 ± 0.01	53 ± 1	264 ± 16
Permeate	0.21 ± 0.02	51 ± 1	239 ± 22
Whey	0.30 ± 0.02	51 ± 1	171 ± 12
Permeate	0.27 ± 0.02	57 ± 1	217 ± 15

TABLE 4.1.26
Percentage Transfer of Cs-137 from Starting Product to End Product

Pathway	Cs-137 % transfer	
	Mean	Range
Whole Milk - Skimmilk	93 ± 35	77 - 139
Whole Milk - Cream	65 ± 3	4.8 - 11
Whole Milk - Whole Milk Powder	88 ± 30	69 - 99
Skimmilk - Skimmilk Powder	120 ± 32	104 - 147
Cream - Butter	*13	11 - 17
Cream - Buttermilk	66 ± 17	51 - 85
Cream - Butteroil	<18	-
Skimmilk - Casein	*0.35	0.15 - 0.60
Skimmilk - Casein Whey	90 ± 25	65 - 110
Whole Milk - Cheese	6.7 ± 2.2	2.7 - 10.5
Whole Milk - Cheese Whey	91 ± 33	84 - 109
Whey - Whey Powder	76 ± 24	47 - 119
Whey - Demineralized Whey	7.4 ± 2.1	5 - 10
Whey - Demineralized Whey Powder	2.3 ± 0.6	0.5 - 4.5
Whey - Rennet Mother Liquor (RML)	51 ± 16	32 - 60
Whey - Delactosed Whey Powder (DLW)	72 ± 15	52 - 84
Whey - Lactose	1.3 ± 0.4	0.4 - 2.9
Whey - Partially Demineralized Delactosed Whey (PDDW)	8.0 ± 0.7	4.4 - 11
Whey - PDDW Powder	33 ± 3	22 - 50

*Calculated using K-40/Cs-137 ratios for products in the same pathway (see Tables 29 and 30)

TABLE 4.1.27
Percentage Transfer of Cs-137 from Whole Milk to Milk Products
 (See Fig. 7)

Milk Product	Percentage Transfer of Caesium-137
Whole Milk Powder	83 ± 30
Skimmilk	93 ± 35
Cream	6.5 ± 2.7
Cheese	6.7 ± 2.2
Cheese Whey	91 ± 33
Skimmilk Powder	111 ± 52
Casein	0.32
Casein Whey	83 ± 39
Whey Powder	66 ± 38
Butter	0.85
Buttermilk	4.3 ± 2.1
Butteroil	<1.2
Demineralized Whey	6.4 ± 3.5
Demineralized Whey Powder	2.0 ± 1.0
Delactosed Whey Powder	63 ± 32
Partially Demineralized Delactosed Powder	29 ± 14
Lactose	1.1 ± 0.6

TABLE 4.1.28
Percentage Distribution of Radiocaesium to Different Dairy Products

Product	Literature Review (Range)	Present Work
Whole Milk	100	100
Cream	3 - 16	6.5
Skimmilk	85 - 99	93
Butter	0.3 - 2.2	0.85
Buttermilk	2 - 13	4.3
Cheese	4 - 23	6.7
Cheese Whey	74 - 96	91
Rennet Casein	1.8 - 7.7	0.32
Rennet Casein Whey	77 - 88	83
Demineralized Whey (90%)	<1.5	6.4 ^(a)

^(a) The difference in the percentage transfer factors for demineralized whey may be attributed to the fact that the products discussed in the literature were manufactured on a laboratory scale unlike those in this study which were collected directly from the creamery.

TABLE 4.1.29

Calculated Cs-137 Concentrations in Butter and the Percentage Cs-137 Transfer from Cream
(Based on the Average K-40/Cs-137 Ratio for Cream and Buttermilk in the same Pathway)

Product	Cs-137 (Bq/kg) or (Bq/l)	Calculated % Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40/Cs-137
Cream	0.46 ± 0.05	12	18 ± 1	40 ± 5
Buttermilk	0.46 ± 0.11		31 ± 2	67 ± 17
Butter	**0.12		6.6 ± 0.8	*54
Cream	0.28 ± 0.04	17	24 ± 1	84 ± 14
Buttermilk	0.47 ± 0.05		30 ± 2	64 ± 7
Butter	**0.10		7.7 ± 1	*74
Cream	0.15 ± 0.04	11	24 ± 1	156 ± 39
Buttermilk	0.26 ± 0.05		44 ± 1	168 ± 35
Butter	**0.04		5.8 ± 0.6	*160

* average K-40, Cs-137 ratio based on ratio for other products in the same pathway

** calculated Cs-137 activity based on assumed K-40, Cs-137 ratio and measured K-40 value

TABLE 4.1.30

Calculated Cs-137 Concentrations in Casein and the Percentage Cs-137 Transfer from Skimmilk
(Based on the Average K-40/Cs-137 Ratio for Skimmilk and Casein Whey in the Same Pathway)

Product	Cs-137 (Bq/kg) or (Bq/l)	Calculated % Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40/ Cs-137
Skimmilk	1.0 ± 0.1	0.15	34 ± 2	34 ± 3
Casein Whey	0.69 ± 0.06		41 ± 2	59 ± 6
Casein	**0.05		2.4 ± 0.6	*47
Skimmilk	0.55 ± 0.23	0.60	68 ± 1	124 ± 51
Casein Whey	0.39 ± 0.08		47 ± 1	120 ± 28
Casein	**0.11		13 ± 1	*120
Skimmilk	0.26 ± 0.03	0.38	43 ± 1	165 ± 20
Casein Whey	0.27 ± 0.05		42 ± 1	155 ± 28
Casein	**0.03		5.4 ± 0.5	*160
Skimmilk	0.24 ± 0.02	0.30	47 ± 1	197 ± 18
Casein Whey	0.28 ± 0.08		49 ± 1	175 ± 49
Casein	**0.03		4.8 ± 1.5	*185
Skimmilk	0.24 ± 0.06	0.30	46 ± 1	190 ± 49
Casein Whey	0.28 ± 0.05		43 ± 1	154 ± 29
Casein	**0.03		4.3 ± 0.6	*170

* average K-40/ Cs-137 ratio based on ratio for other products in the same pathway

** calculated Cs-137 activity based on assumed K-40/ Cs-137 ratio and measured K-40 value

TABLE 4.1.31

Calculated Cs-137 Concentrations in Cheese and the Percentage Cs-137 Transfer from Whole Milk
(Based on the Average K-40/Cs-137 ratio for Whole Milk and Cheese Whey in the Same Pathway)

Product	Cs-137 (Bq/kg) or (Bq/l)	Calculated % Cs-137 transfer	K-40 (Bq/kg) or (Bq/l)	K-40/Cs-137
Whole Milk	0.63 ± 0.09	7	40 ± 3	63 ± 10
Cheese Whey	0.60 ± 0.02		41 ± 2	68 ± 3
Cheese	**0.46		30 ± 2	*65
Whole Milk	0.29 ± 0.02	4	43 ± 2	148 ± 13
Cheese Whey	0.26 ± 0.09		43 ± 1	165 ± 83
Cheese	**0.13		20 ± 2	*157
Whole Milk	0.26 ± 0.07	5	46 ± 1	176 ± 46
Cheese Whey	0.25 ± 0.05		45 ± 1	181 ± 33
Cheese	**0.13		23 ± 1	*178
Whole Milk	0.25 ± 0.07	3	45 ± 1	179 ± 50
Cheese Whey	0.16 ± 0.66		32 ± 1	200 ± 76
Cheese	0.09		16 ± 2	190

* average K-40/Cs-137 ratio based on ratio for other products in the same pathway

** calculated Cs-137 activity based on assumed K-40/Cs-137 ratio and measured K-40 value

TABLE 4.1.32

Calculated Cs-137 Concentrations in Demineralized Whey and the Percentage Cs-137
Transfer from Whey using the K-40/Cs-137 Ratio for Whey

Product	Cs-137 (Bq/l)	Calculated Cs-137 transfer	K-40 (Bq/l)	K-40/Cs-137
Whey	0.37 ± 0.06	5.4	46 ± 1	123 ± 18
Demineralized Whey	**0.02		2.4 ± 0.2	*123
Whey	0.29 ± 0.16	6.8	32 ± 2	154 ± 86
Demineralized Whey	**0.002		0.33 ± 0.03	*154

* K-40/Cs-137 ratio for whey in the same pathway

** calculated Cs-137 activity based on assumed K-40/Cs-137 ratio and measured K-40 value

TABLE 4.1.33
Comparison of Specific Activities (Bq/kg) in Different Whey Powders
at Different Times of the Year (See Fig. 8)

Month Collected	Whey Powder	Demineralized Whey Powder	Delactosed Whey Powder	Partially Demineralized Delactosed Whey Powder
June	2.1 ± 0.1	0.26 ± 0.02	8.1 ± 0.5	4.4 ± 0.3
August	—	0.07 ± 0.02	7.2 ± 0.4	3.1 ± 0.1
November	0.73 ± 0.04	—	2.5 ± 0.1	2.1 ± 0.1

TABLE 4.1.34
Normalized Cs-137 Specific Activities (Bq/kg) in Milk Products
(See Fig. 9)

Whole Milk	1.0
Whole Milk Powder	7.0
Skimmilk	1.0
Skimmilk Powder	14
Cream	0.72
Butter	0.86
Buttermilk	0.95
Butteroil	<0.27
Cheese	1.2
Cheese Whey	0.96
Casein	<0.89
Cheese Whey	0.89
Whey Powder	12
Demineralized Whey	0.09
Demineralized Whey Powder	0.47
Rennet Mother Liquor	8.7
Delactosed Whey Powder	23
Lactose	0.41
Partially Demineralized Delactosed Whey	1.3
Partially Demineralized Delactosed Whey Powder	11

TABLE 4.1.35

Transfer of Radiostrontium and Radiocaesium from Whole Milk to Milk Products [9]

Product	Weight of starting material	Weight of product	Product moisture content	Activity in starting material	Activity in product	Transferred from milk
	kg	kg	%	c/m/g	c/m g	%
Radiostrontium						
Butter	0.55	0.29	31	17.6	1.2	0.06
Cheddar Cheese	6.97	0.50	39	24.3	166	49
Cottage Cheese	3.36	0.27	78	26.2	9	2.7
Radiocaesium						
Butter	0.52	0.17	13	153	24	0.03
Cheddar Cheese	5.81	0.64	44	201	141	7.7
Cottage Cheese	5.21	0.36	56	216	35	1.1

TABLE 4.1.36

Radiostrontium and Radiocaesium Intake from Whole Milk and Cheese on the basis of Calcium, Protein and Calorie Requirements[9]

Product	Basis of Substitution					
	Calcium		Protein		Calories	
	Sr-85	Cs-137	Sr-85	Cs-137	Sr-85	Cs-137
Whole Milk	100	100	100	100	100	100
Cheddar Cheese	94	10	100	10	120	12
Cottage Cheese	89	33	8	3	30	10

TABLE 4.1.37
Maximum Permitted Radioactivity Levels for Foodstuffs and Feedingstuffs (Bq kg)
[66, 71, 72]

Radionuclide	Baby Foods (1)	Dairy Produce (2) (3)	Other Foodstuffs except Minor foodstuffs (4)	Liquid Foodstuffs (5)	Feeding Stuffs
Isotope of strontium, notably Sr-90	75	125	750	125	(7)
Isotope of iodine, notably I-131	150	500	2000	500	(7)
Alpha-emitting isotopes of plutonium and transplutonium elements, notably Pu-239, Am-241	1	20	80	20	(7)
All other nuclides of half-life greater than 10 days, notably Cs-134, Cs-137 (6)	400	1000	1250	1000	pigs 1250 (8) 2500 other 5000

- (1) Baby foods are defined as those foodstuffs intended for the feeding of infants during the first four to six months of life, which meet, in themselves, the nutritional requirements of this category of person and are put up for retail sale in packages which are clearly identified and labelled 'food preparation for infants'.
- (2) Dairy produce is defined as milk falling within headings Nos 04.01 and 04.02 of the Commons Customs Tariff, and the corresponding headings of the combined nomenclature as from 1 January 1988.
- (3) The level applicable to concentrated or dried products shall be calculated on the basis of the reconstituted products as ready for consumption.
- (4) Minor foodstuffs and the corresponding levels to be applied to them will be as defined in accordance with Article 7.
- (5) Liquid foodstuffs as defined by Chapters 20 and 22 of the Commons Customs Tariff and by the corresponding Chapter of the combined nomenclature as from 1 January 1988. Values are calculated taking into account consumption of tap-water and the same values should be applied to drinking water supplies at the discretion of competent authorities in Member States.
- (6) Carbon 14 and tritium are not included in this group.
- (7) For these radionuclides no maximum permitted levels have been laid down because of the low animal transfer factors of the strontium and alpha-emitting isotopes and the short-half-life of I-131.
- (8) Poultry, lambs and calves.

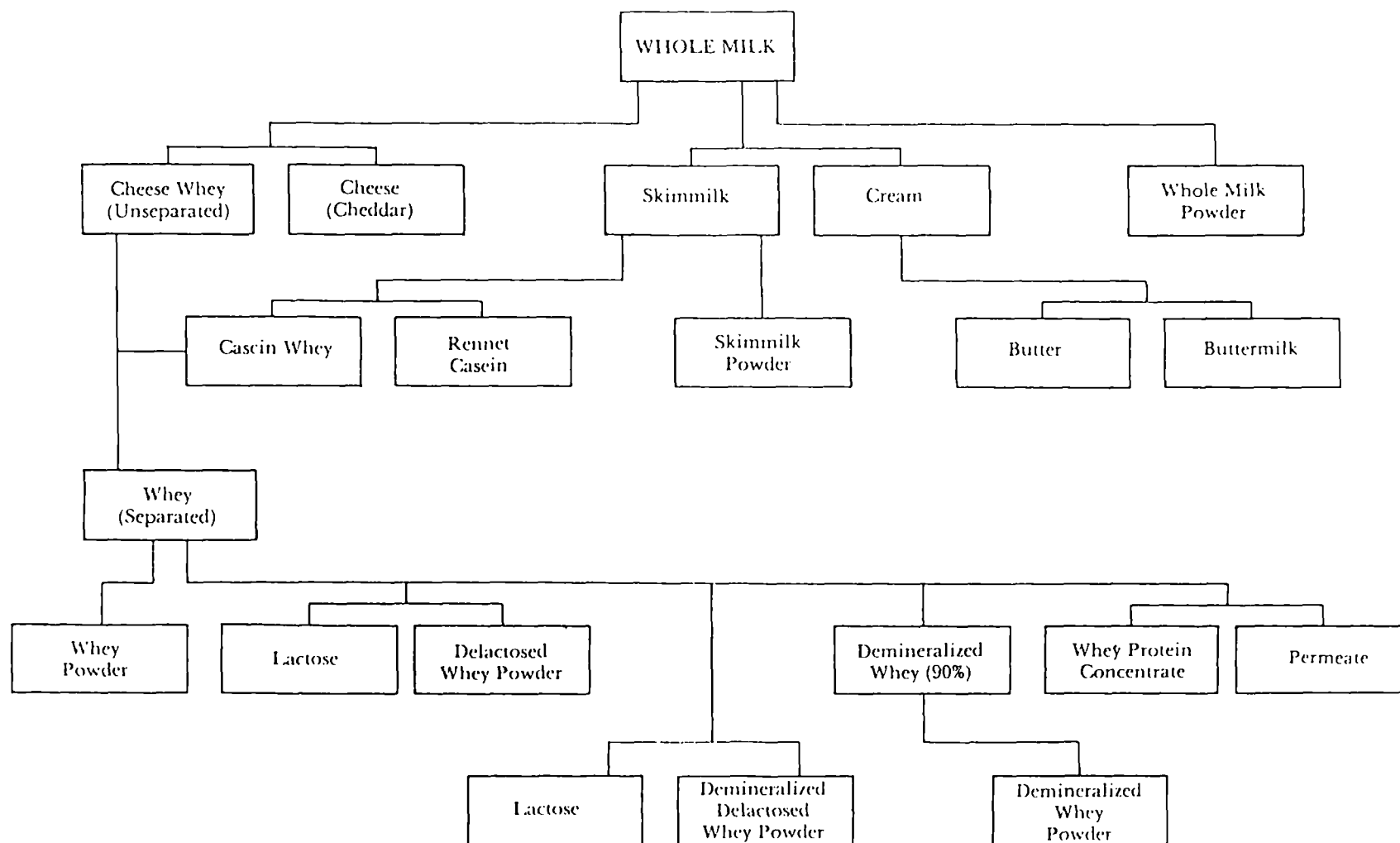


FIG. 4.1.1
Principal Milk Processing Pathways

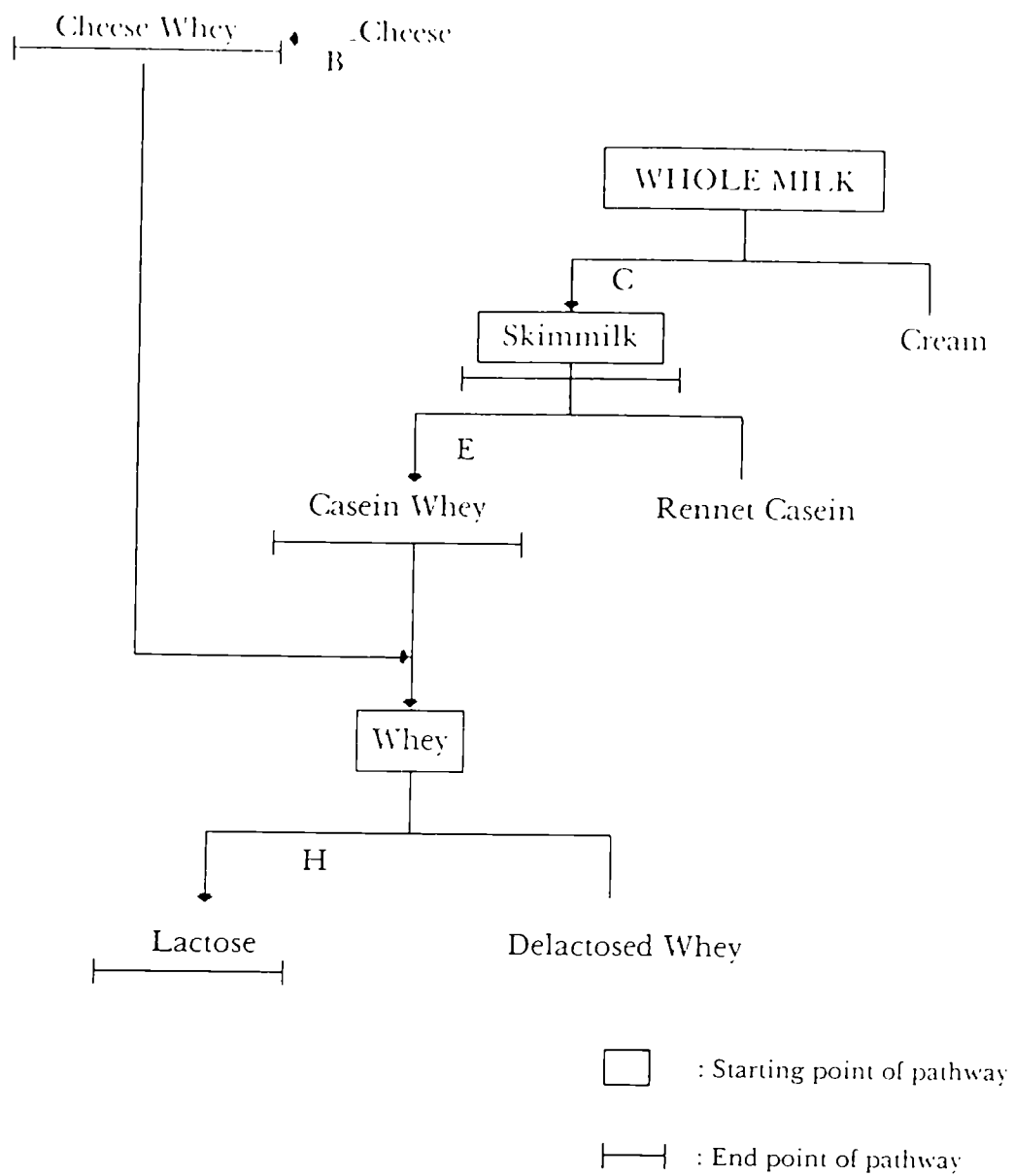


FIG. 4.1.2
Whole Milk to Lactose Pathway

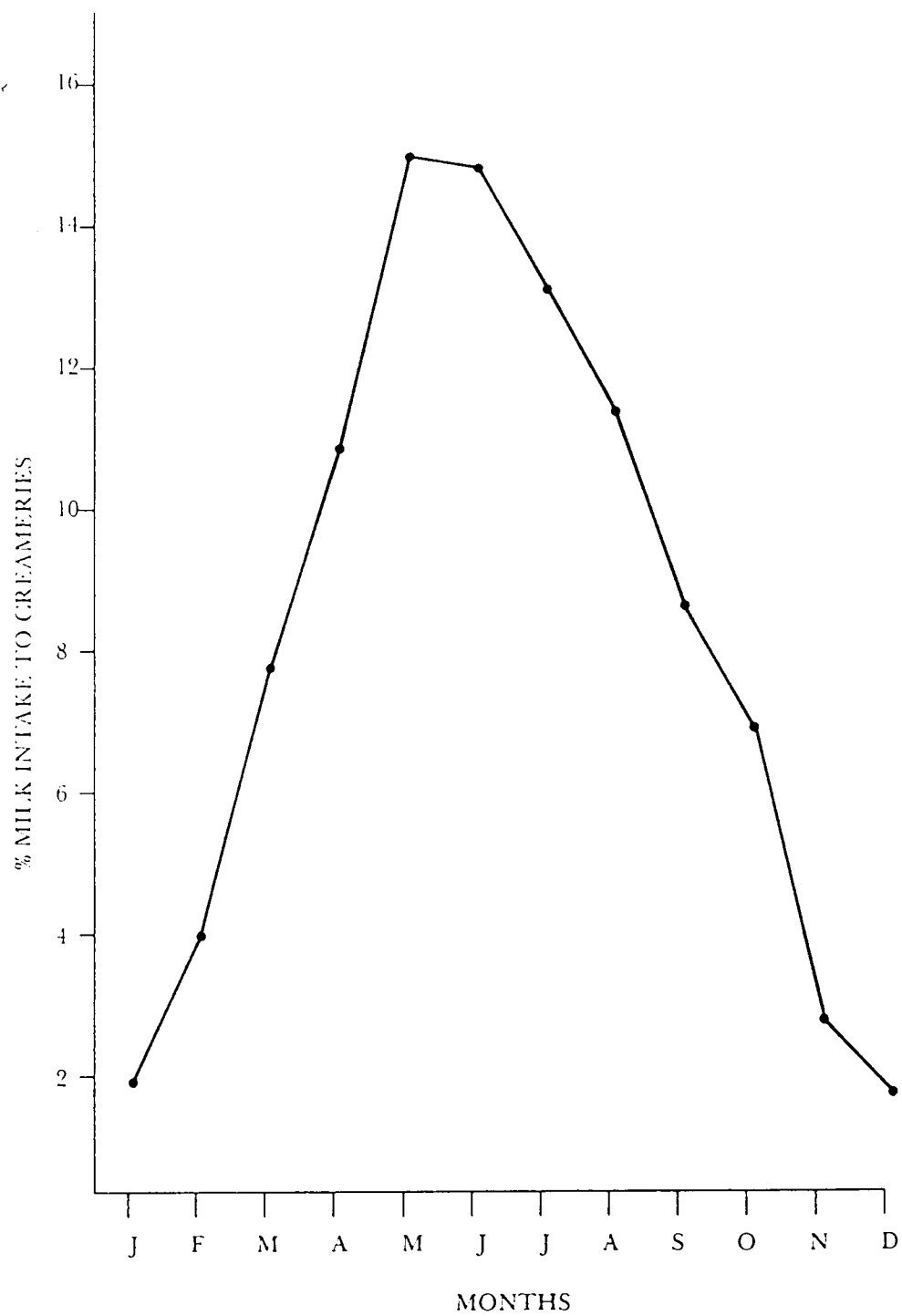


FIG. 4.1.3
Typical Milk Production Cycle in Ireland

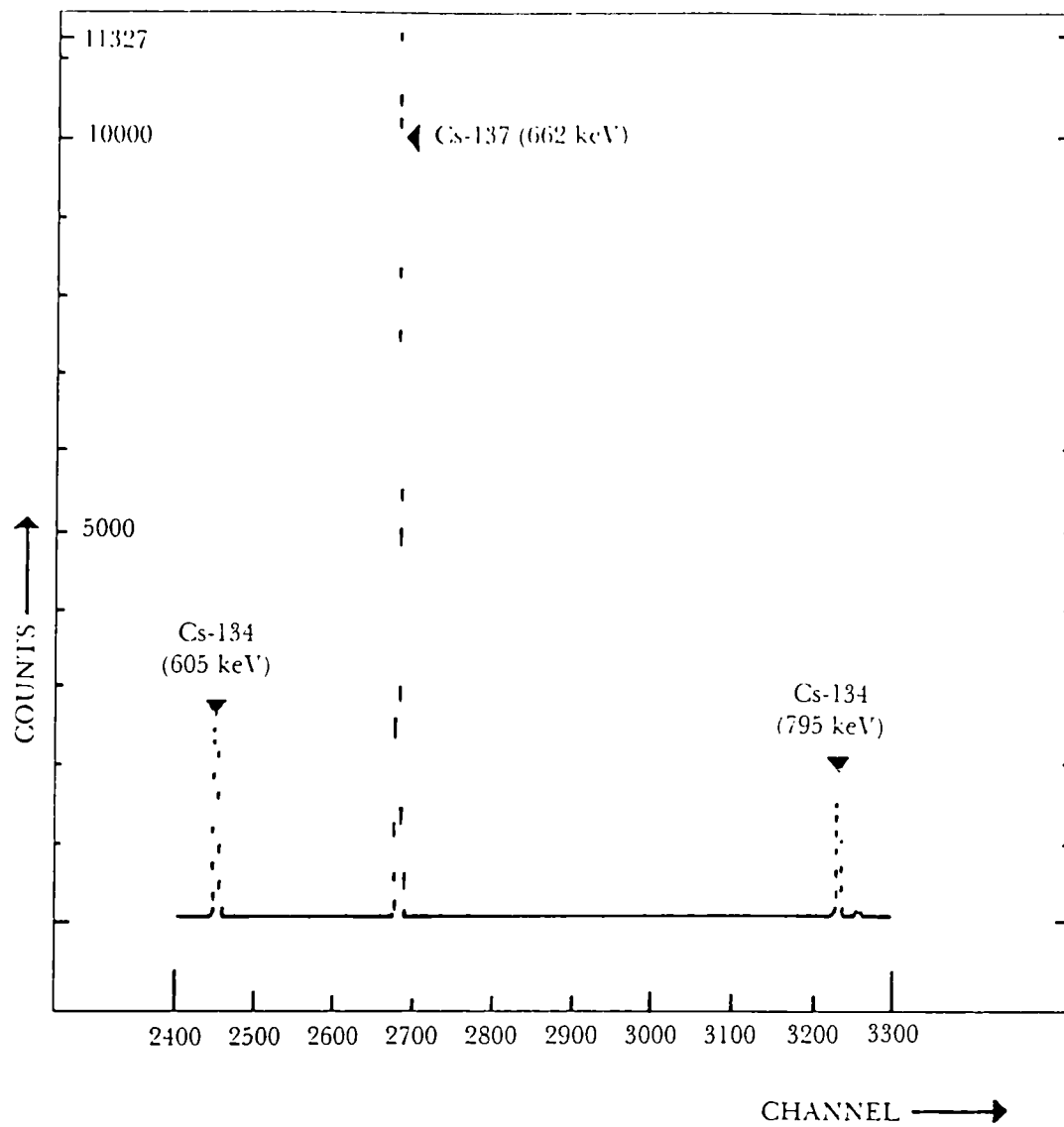


FIG. 4.1.4
Typical (expanded) Milk Powder Spectrum
(Sample Mass, 271.55 (g); Counting Time 9913 (s))

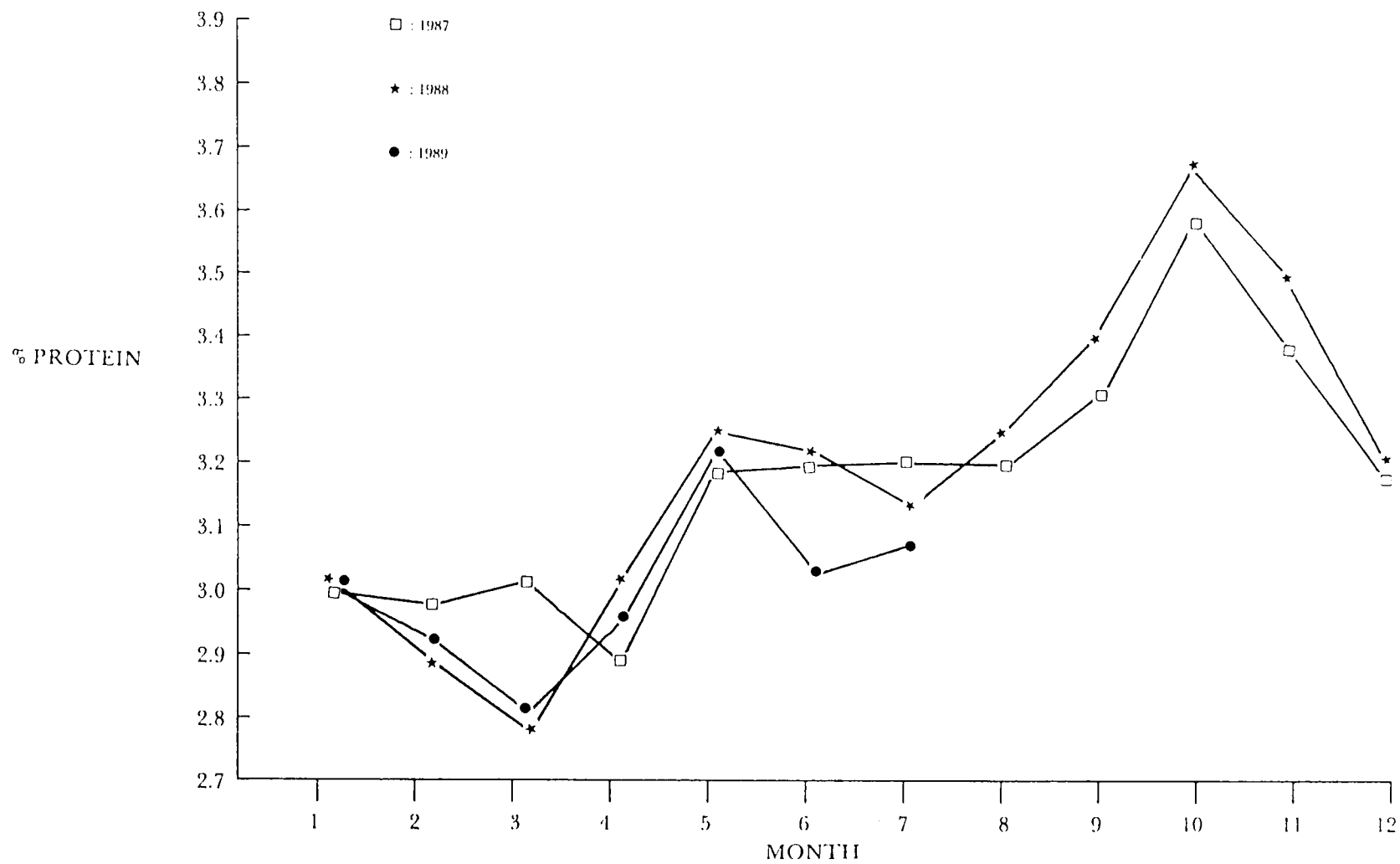


FIG. 4.1.5
Seasonal Variation of the Protein Content in Milk

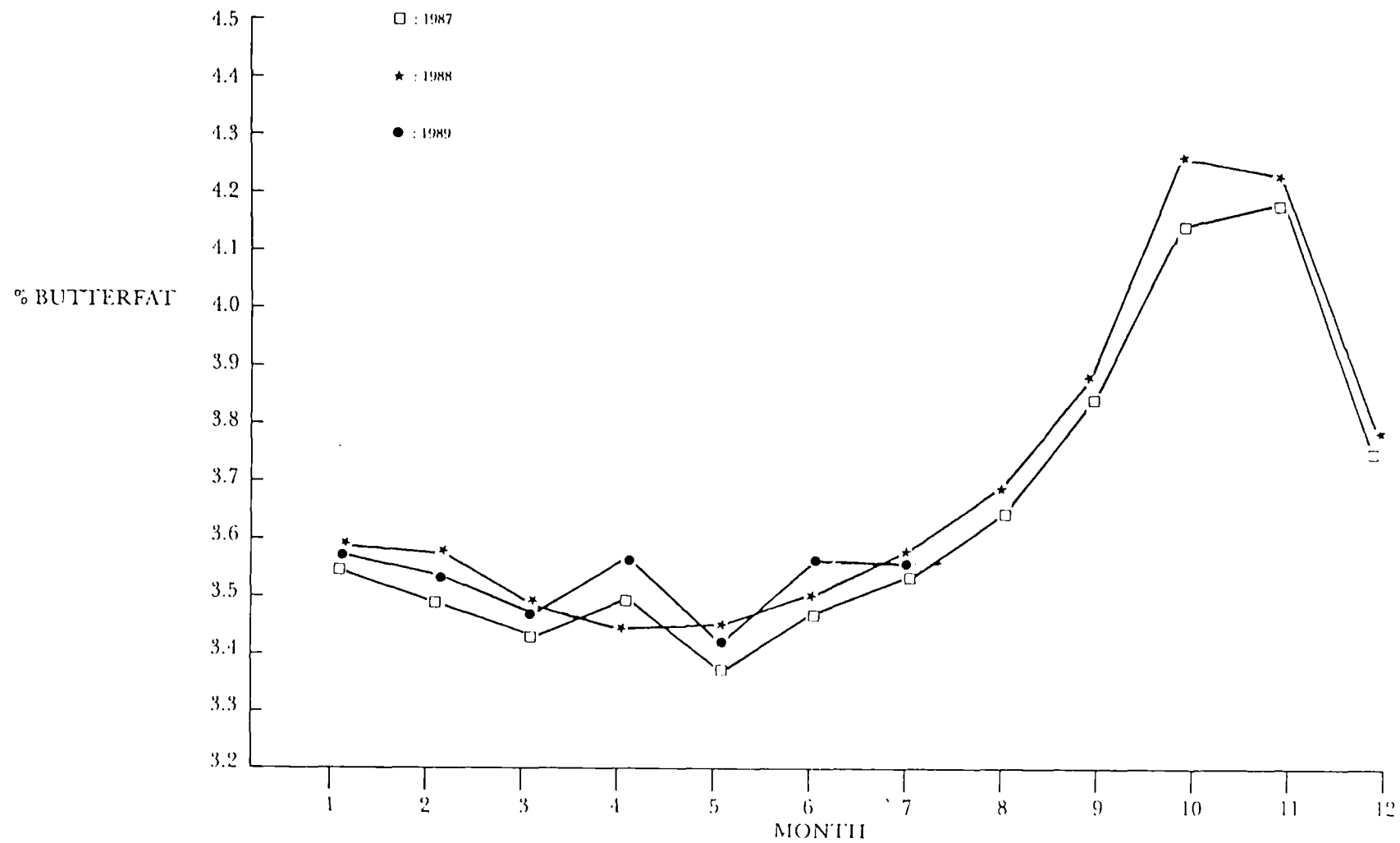
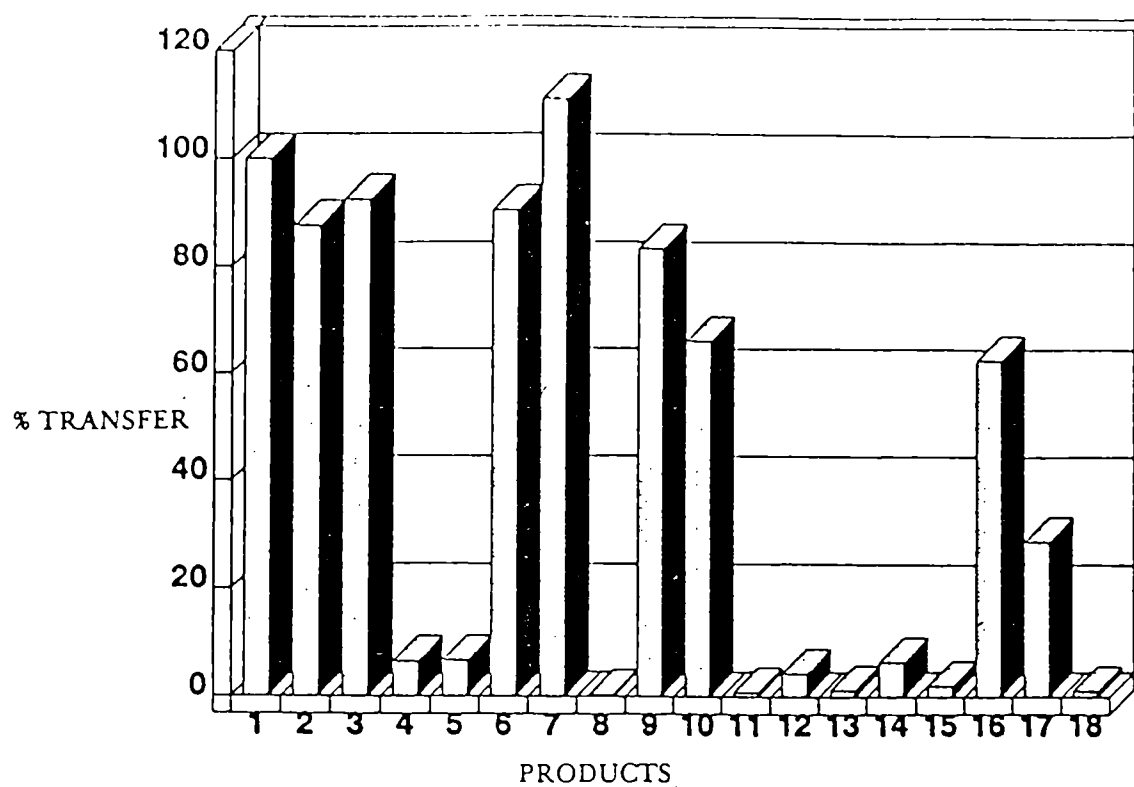


FIG. 4.1.6
Seasonal Variation of the Fat Content in Milk



- | | |
|----------------------|---|
| 1. Whole milk | 10. Whey powder |
| 2. Whole milk powder | 11. Butter |
| 3. Skimmilk | 12. Buttermilk |
| 4. Cream | 13. Butteroil |
| 5. Cheese | 14. Demineralized whey |
| 6. Cheese whey | 15. Damin. whey powder |
| 7. Skimmilk powder | 16. Delactosed whey powder |
| 8. Casein | 17. Partially damin. delac. whey powder |
| 9. Casein whey | 18. Lactose |

FIG. 4.1.7
Percentage Transfer of Caesium from Whole Milk to Milk Products

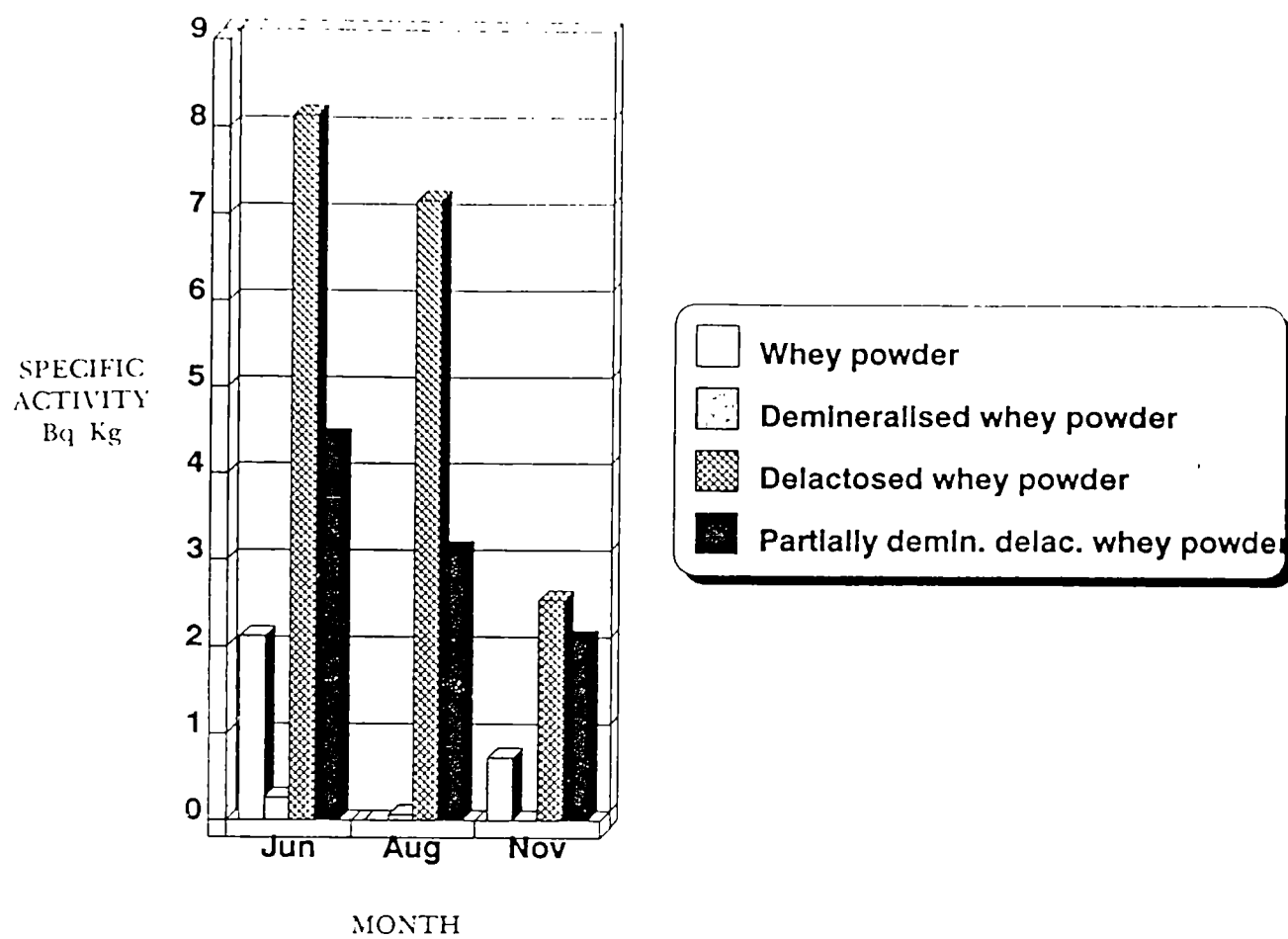
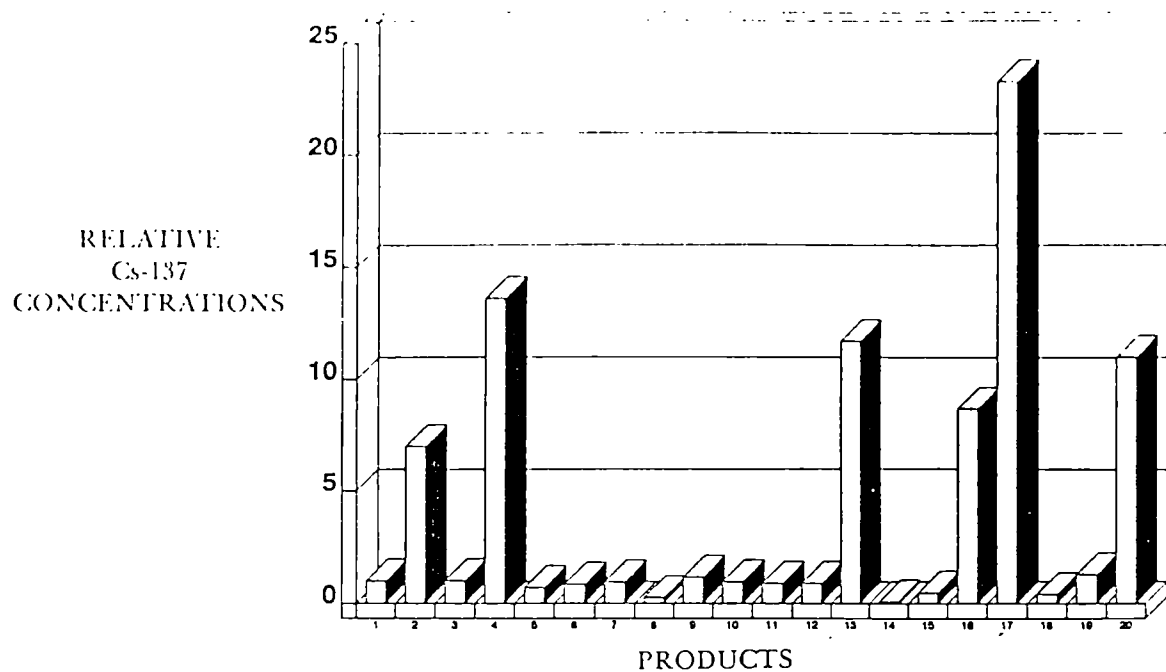


FIG. 4.1.8
Cs-137 Concentrations in Different Whey Powders



- | | |
|----------------------|---|
| 1. Whole milk | 11. Casein |
| 2. Whole milk powder | 12. Casein whey |
| 3. Skimmilk | 13. Whey powder |
| 4. Skimmilk powder | 14. Demineralized whey |
| 5. Cream | 15. Demin. whey powder |
| 6. Butter | 16. Rennet mother liquor |
| 7. Buttermilk | 17. Delactosed whey powder |
| 8. Butteroil | 18. Lactose |
| 9. Cheese | 19. Partially demin. delac. whey |
| 10. Cheese whey | 20. Partially demin. delac. whey powder |

FIG. 4.1.9
Normalized Cs-137 Specific Activities in Milk Products

5. DISCUSSION

E. IRANZO and G. ARAPIS

INSTITUTO PRYMA - CENTRO DE INVESTIGACIONES ENERGETICAS MEDIOAMBIENTALES Y
TECNOLOGICAS - MADRID (SPAIN)

It is quite clear that reclamation of rural areas after accidents, with some especial exceptions, had not been given much attention in the pre-Chernobyl era. Nevertheless, contamination of some rural areas in or near the site of a nuclear accident, a nuclear test or a high radioactivity release have posed problems of reclamation and of research on this subject, from which the Community could learn. The research has been very limited so far, but direct information on countermeasures taken and research in some related fields, such as soil-plant transfer factors and radionuclide transference in food processing, could help for recommending decisions, designing some action model and establish the pathways of needed research. This information could be used for selecting, in the near future, a scale of countermeasures techniques to face the different kinds and levels of radioactive contamination derived from an accident at nuclear installation.

With that purpose, the task of the collaborating laboratories in the development of this action has been directed to:

1. Collect and elaborate the available information about:

- Deposition of contamination on vegetation and soils (direct contamination).
- Behaviour of contaminants in the soil and transfer into the plants (indirect contamination).
- Reduction of the contamination levels in the rural environment by:
 - Removal of contaminated vegetation and agricultural products.
 - Application of agro-technical countermeasures to soils.
- Reduction of the radiological impact to people by:
 - Food processing.
 - Selection of crops and soil uses.
- Follow-up of the effectiveness of the undertaken countermeasures in rural areas.

2. Start some supplementary investigations on:

- Acquisition of data related to the deposition of radionuclides and specially evaluate their interception rate by the vegetative surface of several crops at different growth states.
- Possibilities to diminish the soil-plant transfer of contaminants by using mineral elements.
- Transfer factors for different species of crops growing on several types of soil.
- Study of the transfer of radiocesium from raw whole milk to a wide range of milk products produced by the dairy industry.

The main conclusions achieved during this Action by the laboratories of CEA (Cadarsache), CIEMAT (Madrid), NEB (Dublin), RNL (Risø) are discussed below, according the following natural sequence of activities that would be considered in order to establish actions in the case of an accident:

- Historical analysis of the land recovery operations.
- Characteristics of contamination:
 - Deposition study of the contaminants over the vegetation cover.
 - Behaviour of the radionuclides in the soil and their transfer to vegetation and crops.
- Countermeasures applicable:
 - Treatment of vegetation.
 - Treatment of contaminated soils (mechanical and chemical).
 - Alternatives to these treatments (crops selection and food processing).
- Evaluation of the countermeasures to be undertaken.

Finally, some consideration about further needs, in relation with the CEC Action, will be pointed out.

5.1 THE PAST HISTORY OF REMEDIAL ACTION

An extensive analysis of the scenarios where an accident has taken place, a nuclear test has taken place or a high radioactive contamination was produced with the consequent impact in the agricultural environment, was made in Spain (CIEMAT). The reviewed scenarios were:

- Accidents: Windscale (UK, 1957), Palomares (Spain, 1966), Thule (Denmark, 1968), Rocky Flats Plant (USA, 1958) and Chernobyl (URSS, 1986).
- Nuclear tests: Bikini Atoll (1946-1958), Enewetak Atoll (1948- 1958) and Nevada Test Site (1951-1975) (USA).
- Others: Hiroshima and Nagasaki (Japan, 1945), and Los Alamos (USA, 1942-1965).

Information about the type and use of soil, kind and level of contamination, cleanup criteria and countermeasures applied per scenario, is presented in the table 5.1. The information about laboratory and other field researches on countermeasures for land recovery was, also, compiled and a summary of them is included in table 5.2.

The analysis of this information shows important details about countermeasures' application per scenario which, in the past, have been used. Interesting information was, also, obtained which facilitates the final evaluation of the recovery operations, below presented.

A Danish (Riso) review of literature (ref. 1 of chapter 3.3) had the aim of collecting knowledge on root uptake of cesium, strontium and plutonium that makes it possible to estimate the applicability of different treatments in land reclamation in distant areas after release of these radionuclides. The main conclusions of the behaviour of these contaminants in the soil-plant ecosystem are presented below in paragraph 5.2.2.

5.2 THE CONTAMINATION

In connection with accidents in nuclear facilities, few data were available. The majority of the studies on environmental radioactive contamination concerns the weapons fallout from the nuclear tests period 1954-1965, and they are focussed on strontium-90, caesium-137 and plutonium-239, radionuclides that constituted the principal internal hazard arising from that fallout.

Table 5.1 Scenario's description, cleanup criteria and countermeasures

SCENARIO	USE	SOIL TYPE	CONTAMINANT	LEVEL OF CONTAMINATION	CLEANUP CRITERION	COUNTERMEASURE
Los Alamos	Residence Recreation activities	Desert and soft volcanic rock	U, Pu and fission products	Pu ²³⁹ : 8.2 kBq/kg Cs ¹³⁷ : 4.6 kBq/kg	ALARA	Soil removal
Bikini	Residence and agricultural	Coral rock and coralline sands with minimum organic content	Transuranics Cs, Sb, Co, Rh	< 120 µR/hr	< 5.92 kBq/kg averaged over 1/16 ha (for soil removal)	Decay Debris removal Devegetation Soil removal
Enewetak			Transuranics Cs, Sr, Co	Pu ²³⁹ <20. kBq/kg Cs ¹³⁷ <6.7 " Cs ⁹⁰ <25. " Sr ²⁴¹ <1.2 " Am		Devegetation Plowing Soil removal
Nevada	Desert	Sandy Calcareous alluvium	Reactor fission and activation products	300 Acres greater than 0.37 MBq/m ² Few acres greater than 2.6 MBq/m ²	< 5.92 kBq/kg averaged over 1/16 ha (for soil removal) and ALARA	Plowing Soil removal Vacuum Scraping Oiling Leaching
Windscale	Urban and agricultural		I ¹³¹ Cs ¹³⁷ Cs ⁹⁰ Sr ⁸⁹ Sr	7.4 10 ¹⁴ Bq total 2.2 10 ¹³ Bq " 3.0 10 ¹² Bq " 3.3 10 ¹¹ Bq " < 4mR/hr	Milk samples < 0.1 µCi/l Decrease activity with half life < 8 days	Milk consumption restriction

Scenario's description, cleanup criteria and countermeasures
(Table 5.1, continuation)

SCENARIO	USE	SOIL TYPE	CONTAMINANT	LEVEL OF CONTAMINATION	CLEANUP CRITERION	COUNTERMEASURE
Rocky Flats Plant		Very rocky 20% clay & organic matter	Pu	200-30000 kBq/m ² in drum storage area; in Lip Area 20.7 GBq	Soil removed if > 1000Bq/g and returned if < 30 Bq/g	Removal of soil Stabilizaters
Palomares	Urban and agricul- tural	Silt and sandy	Pu	>1200 kBq/m ² 2.2 ha 120-1200 17 " 12-120 87 " <12 120 "	> 562 µg/m ² 5.4-562 " < 5.4 " < 77 "	*scraped + removed *water + plow *water *permissible where other measures could not be appllied
Thule		Ice	Pu	≤ 14.8 GBq/km ²		Ice removal
Chernobyl	Urban and agricul- tural	Sandy and argillo- arenaceus sward- podzolic acid	Cs ¹³⁷ and fission products 2x10 ¹⁸ Bq released	50% of released material in 30 km zone	To recover the area (levels ?)	Devegetation Soil removal Plowing Stabilizers Fertilizers

Table 5.2 Laboratory and field research studies

COUNTERMEASURE	RESEARCH CENTER	SUBJECT
Removal of vegetation	Agricultural Research Service, Beltsville, Md.	Comparison of methods for removal
Removal of soils	Nevada Test Site (N.T.S.) US Department of Defense US Army Nuclear Def. Lab. AWC. Inc., Las Vegas, Nevada	Test the vacuum methodology Effectiveness of machinery and methodologies Comparison of methods for removal in normal and cold weather conditions Development of a soil decontamination plant
Ploughing	N.T.S.	Effectiveness of normal ploughing with chemical additives
Deep ploughing	Agricultural Research Service, Beltsville, Md.	Comparison of strontium uptake to different depths
Leaching	N.T.S.	Effectiveness by water and chemical solutions
Application of soil amendments	CEA-Cadarache, Chernobyl and others	Specific reduction of radionuclides uptake
Application of surface fixatives	Pacific Northwest Lab. Chernobyl N.T.S. Rocky Flats Plant	Effectiveness and cost studies Feasibility of foam application Effectiveness of oil application Effectiveness of asphalt and foam application
Physico-Chemical treatment of soils	Rocky Flats Plant	Feasibility and effectiveness of several methods

In the case of a nuclear plant accident a large number of nuclides with distinct nature or chemical form are dispersed and, thus, the consequent radiation risk differs from that of tests' fallout. In fact, the role of strontium in a plant accident scenario is minor. However, its contribution to establish our actual knowledge, about the radionuclides behaviour in the system "soil-plant", is great.

5.2.1 DEPOSITION OF CONTAMINATION

The qualification of the contamination repartition between soil and plant could be used to decide the countermeasure to be applied in the soil reclamation process. One of the possible countermeasures is the removal of vegetation, and it could be the unique one or the first step for further decontamination actions. The recovery efficiency depends on the machinery used when any countermeasure is applied.

The percentage of repartition between the vegetation and soil depends on the kind of vegetation and the growth state of plants at the moment of the deposition.

Some data about this subject could be obtained by reviewing past scenarios, but its high interest in the planning of future countermeasures was considered in CEA-Cadarache to develop an important experimental study (chapter 3.1). So, the investigations made using an extremely trusted uranine labeling method, simulating an accidental dry deposition, have given more data precise than in the past and more dependent on meteorological conditions.

The aim of this work has been the study of contamination repartition between land and vegetative cover for the different kind of crops (meadow, wheat, maize, sunflower, tomato, green peas, bean, colza, cabbage, Brussels sprouts, potato, green salad, beetroot, lucerne and linen) and in function of their vegetative and sanitary state, and their density.

The curves of the interception coefficient because of the aerian part of the vegetative productivity (g dry/m^2), elaborated for potato, wheat, sunflower and maize, show an important variability. These results indicate the needs to correct our actual knowledge about deposition velocity in function of the kind of vegetation. It seems that the consideration of a unique deposition velocity which is related with a concentration in volume (air) and a deposition on surface (soil) is not enough for the general description of the pollutants' deposition. For more realistic interpretations it would be necessary to take in consideration a "surface's equivalent" due to the kind of vegetal cover or to determine a specific deposition velocity related directly with each distinct crop and/or his vegetative step and state of growth.

The mathematical relations proposed for some of the crops studied are as follows:

$$\begin{aligned}\text{Wheat: } y &= 42.94 + 0.11x - (8.2\text{E-}5)x^2 \\ \text{Corn: } y &= 26.71 + 0.14x - (6.43\text{E-}5)x^2 \\ \text{Sunflowers: } y &= 25.63 + 0.28x - (4.32\text{E-}4)x^2 \\ \text{Potatoes: } y &= 14.95 + 0.56x - 0.001x^2\end{aligned}$$

The table 5.3 shows the rates of contamination deposited on crops.

Table 5.3 Rates of contamination deposited on crops

PLANTS WITH IMPORTANT VERTICAL GROWTH
(CEREALS, MAIZ, SUNFLOWER)

POST GERMINATION VEGETATIVE STATE	20% - 50%
CELLULAR ELONGATION'S STATE	40% - 70%
FRUCTIFICATION'S STATE	65% - 90%
FRUIT MATURITY'S STATE	60% - 85%

MARKET-GARDENER PLANTS
(LETTUCE, PEAS, GREEN BEANS)

CELLULAR ELONGATION'S STATE	20% - 30%
FRUCTIFICATION'S STATE	20% - 30%
FRUIT MATURITY'S STATE	40%

PLANTS WITH LARGE SURFACE LEAVES
(BEET, CABBAGE)

CELLULAR ELONGATION'S STATE	50%
-----------------------------	-----

5.2.2 CONTAMINANTS IN THE SOIL-PLANT ECOSYSTEM

A complete literature study of the behaviour of Cesium, Strontium and Plutonium in the soil-plant ecosystem was made in the Danish Riso National Laboratory (ref. 1 of chapter 3.3). The main conclusions of this study are summarized in continuation.

Behaviour of contaminants in the soil

In soils, contaminants could exist in the following compartments:

- Free in the interspaces (soil-air and soil-water).
- Absorbed to the surfaces of the soil particles.
- Fixed by micaceous elements or in organic complexes.
- Immobilized by microorganisms.
- Fixed as precipitation on surfaces of soil particles.

An equilibrium exists between these compartments. Normally the contaminant in the first two compartments are available for root uptake contrary to the fixed and immobilized ions, which are released only slowly.

- Fixation in soil

There is some confusion on the application of the term fixation and here it is used only for contaminants in an unavailable form.

Cesium:

Fixation of ^{137}Cs is a time dependent process. There is some evidence that trace amount of Cs often behave differently from macro amounts in the soil:

- When it is added in trace amounts to soils, fixation is the most important process concerning the amount of Cs-137 that would be available to root uptake.

The fixation increases with:

- . Increasing clay content; the clay minerals illite and vermiculit are especially important in uncertaining the fixing capacity.
- . Increasing potassium content, since the non-exchangeable potassium is proportional to the content of clay minerals in the soil.
- . Decreasing organic matter, as the organic matter becomes responsible for the major part of the cation exchange capacity.

The influence of pH and contents of exchangeable calcium on the absorption of Cs-137 by soils is less than those of the above mentioned factors.

The addition of stable Cs and NH_4 ions brings more ^{137}Cs into the soil solution. The stable Cs added to contaminated soils displaces the Cs-137 into the soil solution and in that way makes it more available to plants.

- When ^{137}Cs is added in macro amounts, the fixing capacity of the soil may be exceeded and its cation exchange capacity will be responsible for the retention of Cs-137 by absorption.

Strontium:

The organic matter of soil retains strontium in a way that makes it unavailable to root absorption. Clay soils with high degrees of base saturation, also, results in a low strontium uptake.

Plutonium:

Plutonium is fixed in soil colloids, especially clay minerals as kaolinit and illit, making it less mobile.

- Mobility

The causes of the migration of radionuclides in soil can be clasified into two main groups:

- External factors $\left\{ \begin{array}{l} \text{Naturally occurring (i.e. rain).} \\ \text{Anthropogenous (i.e. tilling).} \end{array} \right.$
- Diffusion, due to differences in the concentrations of the nuclide at various points within the soil.

The movements of contaminats through infiltration can be evaluated by the distributiont coefficient k_d :

$$k_d = \frac{\text{absorbed and fixed contaminant per gram soil}}{\text{disolved contaminant per ml of solution in equilibrium}}$$

If K_d is high, no appreciable amount of contaminant would move into the profile by infiltration, and it's less available to plants.

Cesium:

When litter horizons are present, a substantial part of ^{137}Cs can be expected to be bound in these horizons, so the vertical movement by infiltration or diffusion results to be quite small. But the diffusion plays an appreciable role in migration in the cases of:

1. clayish soils in which the infiltration is low,
2. organically rich soils with a high adsorption capacity,
3. bog soils possessing high and relatively permanent water tables, and
4. soils with small water fluxes.

Depending on the content of micaceous minerals, the retention of ^{137}Cs against vertical migration becomes more or less pronounced.

In undisturbed soils, about 90% of ^{137}Cs present in the soil is retained in the upper 5 cm of the soil profile. When leached with water and solutions of NaCl and of CaCl_2 , some downward migration can be expected.

In tilled soils, ^{137}Cs becomes distributed uniformly through the ploughed layer, normally about 20 cm deep. Beyond this depth, the activity decreases in the same manner as in undisturbed soils. The contaminant distribution is markedly different than in uncultivated soils.

Strontium:

It is generally considered that radiostrontium is more mobile in soil than e.g. radiocaesium, but still for many years at least, it has been concentrated in the upper few-cm soil layers.

Plutonium:

The mobility of plutonium in soil is low, because of fixation to soil colloids, especially clay minerals.

When high mobility in soil is recorded, it is often in connection with mechanical events in the soil surface, e.g. frost/thaw, wet-dry phenomena, or irregularities in the structure of the upper earth layers, ridges, fissures, etc.

Most fallout plutonium is nevertheless found in the upper 10 cm of the soil layer, often in the upper 5 cm, where it is available for rainsplash and resuspension. The standard agricultural effects on plutonium distribution in soil merely serve to mix it into the upper 15-20 cm of soil.

However, little information is available in relation to the problems of the resuspension of the contaminating material, and additional investigations seems necessary.

- Loss of activity from the soil-system

The following processes can account for losses:

- Physical decay
- Erosion

{	Wind erosion	{	Wind resuspension
			Surface runoff
	Water erosion		Infiltration to ground water
			Rain splash
- Mechanical resuspension
- Soil ingestion
- Uptake by crops

Cesium:

In organically rich soils loss of activity through infiltration, runoff, resuspension and other erosion phenomena can be expected to be quite small. Physical decay accounts for the greatest losses.

¹³⁷Cs is retained in the upper few centimeters of the soil and it is exposed to loss from erosion. In mineral soils the losses can reach considerable amounts, especially on unvegetated soils. Erosion is consequently greatest in winter. Determined by the local conditions different processes may account for the greatest losses.

The implication of resuspension is not well known, and it is accepted that more than 10% of the deposited material cannot be redistributed.

Transference of contaminants to plants

Two mechanisms contribute to the contamination of vegetation:

- Foliar or direct contamination.
- Root uptake from soil or indirect contamination.

The foliar contamination is a short-term mechanism, if resuspension is not considered, while root uptake may last several years and could continuously contribute to the radionuclides transfer through the food chains.

- Contaminants' availability to root uptake

For the root uptake of the contaminants, their availability into the soil take an important role.

Cesium:

The availability of ^{137}Cs to plant is time dependent due to uptake by plants and to fixation in soil. It has been found that its fixed fraction in soil vary: 75% for vine loam soil and 85% in Canadian mineral soils.

In the soil-system, it is apparent that the single most important factor working on the available and subsequent uptake, is the fixing capacity, primarily determined by the mineral composition and the content of organic matter and potassium.

The supply of stable Cs and ammonium makes ^{137}Cs more available for uptake, while the mobilization by microorganisms makes it less available.

Strontium:

The percentage of available (i.e. soluble and exchangeable) radiostrontium is generally higher than for radiocesium and plutonium. About 90-100% will still be available to root absorption even after many years.

- Root uptake

The relative order of fission products' uptake is as follows:

$^{89-90}\text{Sr} \gg ^{131}\text{I} > ^{140}\text{Ba} > ^{137}\text{Cs}, ^{106}\text{Ru} > ^{144}\text{Ce}, ^{91}\text{Y}, ^{147}\text{Pm}, ^{95}\text{Zr}-^{95}\text{Nb}$

In general, the plant parameters having influence on root uptake of the contaminants are:

- Plant species: root crops and crops with superficial root system tend to accumulate most Cs-137.
- Root depth, since it's related with the contaminant profile in soil.
- Developing stages and plant growth.
- Time of contamination relative to growth season.

The soil variable that generally decreases the Cs, Sr and Pu uptake is the increase of the clay content.

Cesium:

The Cs uptake is similar to potassium mechanisms. The soil variables factors having an effect on Cs-137 root uptake are:

- Uptake decreases with increasing:
 - Clay content.
 - Potassium content, specially when there is a low initial content; when cropping makes a reduction of soil K an increase of Cs-137 uptake by roots is produced.
 - Calcium content on acidic soils.
 - Phosphate content (slight decrease).
- While the uptake increases with increasing:
 - Organic matter contents.
 - Ammonium; it brings Cs-137 into the soil solution phase.
 - Amount of stable Cs (only in small quantities), because it produces a carrier effect, the time of application is essential for the result.
 - Sulphate contents (slight increase).
 - Soil water contents.

Some treatments were shown to have no or a very slight effect on uptake:

- Na, Li, Ba, Mg and often Ca.
- Chelates.
- Minerals on clay soils.
- Cs-137 from irrigation water.

Strontium:

Soil properties that made strontium root absorption low are:

- High clay concentration, while the effect of clay type seems to be less certain.
- Organic matter, since it retains Sr.
- Addition of lime in acid soils with low calcium, while the effect on uptake is limited in neutral, clay and calcareous soils.
- Soil moisture had little effect on plant contents of calcium and strontium.

The effects of additives in strontium root uptake is as follows:

- Nitrate: Stimulating effect.
- Phosphate: Decrease the uptake.
- Chelates: There is no sensational effects.
- Stable strontium: More total uptake with dilution of radiostrontium.

About uptake of ⁸⁹Sr by different plant species, the lowest concentration was found in the gramineae (grasses and cereals). Appreciably larger contents of Sr-89 was found in most of the crucifers (brassicacae, e.g. cabbage) and the highest concentration occurred in the legumes (clover, pea) and the umbelliferas (e.g. carrot, parsley). Between plant species belonging to the same family there was considerable variation in the uptake of Sr-89,

measured on a dry weight basis. Also, differences in uptake of radiostrontium between varieties of the same species have been observed.

Plutonium:

The following factors influences the plant uptake of plutonium:

- Soil pH: It has a great effect;
pH 2-8 : adsorption 98-100%;
pH 8-12: adsorption 80%.
- Oxidation state: Availability to plants follows the valence order
V > VI = III > IV
- Chelation: They enhance the extrability of soil plutonium and increase plant uptake from soil
- Lime: Decrease the uptake by neutralizing the soil.
- The soil type:
Humus: The Pu is associated with the largest molecules of humus, so it depends on the amount of small molecules that have more mobility.
By increasing organic matter the uptake is reduced.
Clay: It reduces the uptake.
- The differences due to plant species are:
The lichens and mosses have a great ability to concentrate plutonium.
No plant species displays extraordinary high root uptake.

Table 5.4 shows a summary of the soils amendments influence in Cs, Sr and Pu root uptake.

Table 5.4 Influence of soil amendments in root uptake

AMENDMENT	Cs	Sr	Pu
Clay contents	D	D	D
Potassium	D		
Calcium on acidic soils	D		
Phosphate	D	D	
Lime in acid soils with low Ca		D	D
Soil moisture		D	
Organic matter	I	D	D
Ammonium	I		
Cs stable (small quantities)	I		
Sulphate	I		
Soil water	I		
NO ₃		I	
Na, Li, Ba, Mg, Ca	N		
Chelates	N	N	I
Minerals on clay soil	N		
Cs-137 from irrigation water	N		
Stable Sr		DI	
pH low			HA
D = Decrease I = Increase N = Low effect DI = Dilution HA = High adsorption			

- Transfer ratios

A normal way to express the uptake is by the plant-soil transfer ratio or transfer coefficient. The use of transfer ratios in the prediction models for dose-management in agricultural products is possible, but the great variability of the soil conditions limit the usefulness of the ratio in making predictions. In the field measurements a problem is the correction to be made from the direct contamination of plants, including resuspension. This problem may be eliminated in laboratory and greenhouse studies. In pot experiments, however, the experimental conditions contribute significantly to higher transfer ratios. Values of transfer ratios, also, depend on the time between supply and measurement.

Cesium:

An estimate of 0.25 is often used for plant-soil transfer of ^{137}Cs . This value seems to represent the majority of soils and vegetation types although there are great differences and many factors to take into account: The transfer coefficient for grassland is ranged between 0.01 and 0.1 and in forest clearings between 0.05 and 6.

Strontium:

The following average plant-soil ratios, depending on plant part, are reported:

- Tops of root vegetables	10:1
- Leguminous forages and vegetation	7.5:1
- Leafy vegetables, roots of root vegetables, natural vegetation and non leguminous forages	5:1
- Fruits, seeds of leguminous vegetables and forages	1:1
- Potato tubers	0.5:1
- Cereal grains, fruits	0.3:1

These ratios indicate a strontium retaining mechanism that influences translocation within the plant.

Plutonium:

The range of concentration ratios known for plant uptake of plutonium is 10^{-2} - 10^{-8} .

A decrease in plant content of plutonium from root surface to root tissue and from root tissue to shoot is observed, maybe due to a discrimination of plutonium during its translocation.

- Translocation

Cesium:

When introduced into plants ^{137}Cs does not become uniformly distributed throughout the plant. Cesium, in common with monovalent cations, has greater mobility in plants than for example the divalent cation strontium. Cs-137 is translocated readily from the point of introduction.

Strontium:

The observed higher concentration of radiostrontium in root, stem and leaves than in fruits and seeds suggests the existence of a retention mechanism of strontium which influences its transfer to the reproductive parts of the plant.

5.3 COUNTERMEASURES

Once the contamination has been produced and depending on the specific scenario in which the agricultural land is polluted, a repartition of the contamination is done in the different compartments of the media. When the contamination is higher than the intervention levels, remedial action must be decided and developed, using countermeasures, to recover the zone to a normal use. A classification of the different countermeasures that could be applied, and their evaluation through their efficiency and their advantages and disadvantages are presented.

5.3.1 CLASIFICATION

The countermeasures applied to recover the agricultural land can be classified as follows:

- Treatment of vegetative cover that is present at the moment of the accident.
- Chemical treatments of soil:
 - Amendments' application.
 - Herbicides' application.
 - Leaching (with water and/or solution).
 - Stabilizers' application (oils, asphalt, foam ...).
- Mechanical treatment of soil:
 - Removal of a soil layer.
 - Removal of an important part of soil.
 - Normal and deep plowing.
 - Other methods (vacuum, in situ treatment, in cold weather ...).
- Alternatives to in situ soil treatments.

Treatment of vegetation

Only in the cases of Palomares and Chernobyl a removal of vegetation by agricultural harvesting of crops has been referred clearly. Additional information about devegetation is shown in tables 5.1 and 5.9.

The very important problem of disposal of the contaminated vegetative material has received little attention, until now.

Chemical treatments of soil

As it has been shown in the review of the past scenarios, many studies have been made to determine the effects of chemical amendments to soil in the root absorption of several radionuclides. But because its great interest as countermeasure and the lack of some important considerations a special attention has been given at CEA-Cadarache (chapter 3.2.1) to the application of mineral elements to study the possibilities of diminish the soil-plant transfer of contaminants. An experimental work has been developed after a correlations' statistical analysis of the transfer factor and of the amount into the soil of the main elements in the fertilizers. The aim of that investigation has been the study of the influence of the basic fertilizers' elements potassium, phosphorus and nitrogen, on the transfer factor of cesium and strontium, because of the kind of soil and plants. The impact of the application on the soils of potassium, ammonium phosphate or calcium carbonate and hydroxide, and of the foliar spraying of chemicals (i.e. potassium nitrate) was tested.

From that experimental work up to the conclusions that potassium is the only element that can reduce has been obtained 100 times, if the soil has high cationic exchange capacity, the transfer factor of Cs - or to a lesser degree of Sr - and, also, that application of ammonium fertilisers must be proscribed in a case of contamination of agricultural lands by a release of radionuclides has been obtained.

In the soils with a low cationic exchange capacity, an excess addition of potassium produces an increase in the transfer factor.

Mechanical treatment of soil

The total or partial removal of the upper layer of contaminated soil, the normal or deep ploughing and the deep placement into the soil of the contaminants are the main mechanical operations in a case of an accident. Additional information on these treatments is presented on the tables 5.1, 5.17, 5.18, 5.19, 5.20, 5.21, 5.22, 5.23 and 5.24, and in the evaluation of countermeasures made below.

Alternatives to soil treatments

- Choice of crops

Some attention has been given in the past to the substitution of typical crops in a contaminated agricultural area by others with smaller human radiological impact as it has been shown in chapter 2.3.4.4. But as a consequence of the post-Chernobyl Action n° 6, research has been made in Riso N.L. (chapter 3.3) to determine differences in the root absorption of Cs by different species of the same crop in order to use as countermeasure the substitution of species instead of the substitution of the usual crop.

The Danish experimental study concerning the indirect contamination aimed to identify crops with relatively low or high root uptake of radiocesium. Four varieties of barley and three of rye grass have been tested for their sensitivity to Cs uptake from a clay loam and an organic soil, and significant differences have been revealed for the crops of the same species.

The field studies concerning direct contamination from Chernobyl Cs deposition show, also, significant differences between the varieties.

In table 5.5 is presented the uptake of different crop species in clay-loam soils.

Table 5.5 Uptake of different crops species in clay-loam soils

SPECIES	VARIETY	RELATIVE Cs-137	ACTIVITY PER FODDER UNIT(*)	
		GRAIN	STRAW	PASTURE
Barley	Apex	0.90	6.40	
"	Golf	1.00	6.40	
"	Ida	1.20	6.71	
"	Gunnar	1.23	7.39	
"	Anker	1.24	6.35	
"	Sila	1.35	10.23	
Grass	Early perennial			3.38
"	Late perennial			4.48
"	Italian			9.12

(*) 1 fodder unit is the fodder value of 1 kg of barley.

Table 5.6 shows the advantages, disadvantages and further knowledge needed in relation with the choice of crops.

Table 5.6 Choice of crops

ADVANTAGES:

- A significative reduction of the crops contamination could be obtained.
- Requires only the selection of varieties in the species.
- Not change in the farmer uses and techniques is needed.
- Not increase the cost production.
- Not adverse environmental effects.

DISADVANTAGES:

- Not using the straw for fodder.
- Negative economical impact in the total price of the crops.

FURTHER KNOWLEDGE NEEDED:

- Behaviour of varieties of other species in other types of soil.
-

These results indicate that it is posible to reduce the transfer of radionuclides through the food chains by efficient agricultural planning of countermeasures, comprising changes of agricultural practices towards the use of plant species and varieties with low root uptakes.

- Food processing

Some scientific groups have already started investigations to follow the radionuclides' transfer through the food processing to get to know the impact of each derived product in the diet.

Dairy products processing:

An experimental study of the cesium transfer has already started, using the milk processing, in Dublin.

Milk and dairy products constitute a substantial portion of the human diet and they are the principal way by which food-borne radionuclides are ingested.

The Cs-137 and Sr-90 would remain in the human food chain for decades, because their long half-lives, while I-131 will not be present after one month approximately.

The Irish experimental work completes the evidence that more than 80% of the whole milk cesium can be distributed in each of the following products:

- Skim milk.
- Casein whey.
- Cheese whey.
- Cottage cheese whey.

In table 5.7 a literature review shows the % distribution of radiocesium to different dairy products. Table 5.8 presents the relative cesium-137 concentration in the different fractions of these products.

Table 5.7 % Distribution of radiocesium to different dairy products
(literature review)

REFERENCE PRODUCT	Antila et al. (19, 20)	Assimakopoulos et al. (3, 4)	Calapaj & Ongern (13)	Everitt & Paulsson (18)	Kondarakis & Anifantakis (17)	Kantare et al. (20, 21)	Kirchmann et al. (14)	Lagoni et al. (17)	Lengemann (2)	Ongem & Tsugo (9)	Pirhonen et al. (22)	Reavey (10)	Wilson et al. (23)	RANGE
Whole Milk	100	100	100	100	100	100	100	100	100	100	100	100	100	
Cream					3	5.9		16	6	7		6	2.7	3 - 16
Skim milk	94					94		85	94	92		94	99	85 - 99
Butter	0.7		<1.5			0.8		2.2	0.1			<1	0.4	0.1 - 2.2
Buttermilk	5.3					5.1		13					2.3	2 - 13
Butterfat						0		0		0				0
Cheese (Rennet)	11	^a 7.4	<10	5	^c 9		^e 5-23		7.7	9.6	6		4.5	4 - 23
Cheese Whey	89	^b 74					^e 77-96			89	93		93	74 - 96
Sour Cheese	12						12							12
Sour Cheese Whey	88						88							88
Cottage Cheese					^d 4				1.1	4.7		4		1 - 5
Cottage Cheese Whey							73			70		82	95	70 - 95
Acid Casein						4.0	1	1.6						1 - 4
Acid Casein Whey						90	84	83						83 - 90
Acid Whey Proteins						1.0		1.4						1.0 - 1.4
Deproteinized Acid Whey						89		82						82 - 89
Rennet Casein						5.7	7.7	1.8						1.8 - 7.7
Rennet Casein Whey						88	77	83						77 - 88
Rennet Whey Proteins						0.6		2.4						0.6 - 2.4
Deproteinized Rennet Whey						88		80						80 - 88
Demineralized Whey													<1.5	<1.5
Whey Protein Concentrate													6.6	6.6
Permeate													8.8	8.8
Yoghurt (Skim milk)					34									34

^a Gruyere Cheese (Ewe's milk)

^b Whey after removal of cream

^c Refalotryl Cheese

^d Myzithra (Whey Cheese)

^e Different values for different types of cheeses

Table 5.8 Relative cesium-137 concentration in dairy products fractions

Fraction	Cs-137
Whole Milk	1.00
Whole Milk Powder	7.6
Skimilk	1.00
Skimilk Powder	11.2
Cream (40%)	0.55
Butter	0.18
Buttermilk	1.10
Cheddar Cheese	0.48
Cheese Whey (unseparated)	0.99
Rennet Casein	0.14
Casein Whey	0.93
Whey Mix	0.96
Whey Powder	18.1
Delactose Whey Powder	24.0
Lactose	0.47
Demineralized Whey (90%)	0.05
Demineralized Whey Powder	1.9

5.3.2 EVALUATION OF COUNTERMEASURES

A general evaluation of the main countermeasures applicable has been obtained by, mainly, the historical analysis of the scenarios where they were used.

The more important considerations about advantages and disadvantages of the countermeasures to be applied are presented with, also, information about the method used, and the efficiency, cost and practicability, in the form of tables.

5.3.2.1 REMOVAL OF VEGETATION

The residual radioactivity on soil could be reduced by an average of 50%. Its effectiveness depends on the kind and density of vegetative cover, on the growing state of crops, and whether the contamination has a wet or a dry form origin.

The soil layer and its productivity are not disturbed.

The vegetation removal involves the use of special machinery, creating a large amount of waste, produces resuspension during the operations, and has an important environmental impact.

In table 5.9 an efficiency, cost and practicability evaluation of the removal of vegetation is presented.

Table 5.10 shows a comparison of methods for removal of contaminated vegetation, and table 5.11 summarized the advantages, the disadvantages and the further knowledge needed in relation of this countermeasure.

Table 5.9 Evaluation of the removal of vegetation in the scenarios

SCENARIO	METHOD	EFFICIENCY	COST	PRACTICABILITY
Bikini	The vegetative cover was knocked down and left in place to provide additional organic matter for the soil	Associated with turning of soils results in a rapid and good reduction of the measured exposure rate		Problem with the elimination of the organic matter
Enewetak	⇒Two bulldozers at the end of a chain, move in parallel ⇒Pushing vegetation into windrows with a bulldozer ⇒Use of front end loader ⇒Use of road grader with scarifier teeth	⇒Not very optimum ⇒Unsuccessful machine (it does not remove many roots and bush stumps)	⇒The most rapid technique	⇒It depends on the surface cover ⇒Problem: Mixes and spreads the surface soil ⇒Least satisfactory as a means of removing stumps and roots
Palomares	Use of machetes, sickles, scythes hoes and removal	Good	306 m ³ removed and stored and 92.5 Mkg burned of a 1.15 km ² area	Depends on the crop. Real decontamination.

Table 5.10 Comparison of methods for removal of contaminated crops or mulches from land

TYPE OF VEGETATION	IMPLEMENT	REMOVAL OF RADIOACTIVITY %	EFFORT REQUIRED	
			FOR REMOVAL	FOR DISPOSAL
SOYBEANS (12" HIGH)	MOWER	<75	POOR	FAIR
" "	FLAIL HARVESTER	<75	FAIR	GOOD
" (FULL GROWTH)	" "	<75	POOR TO FAIR	GOOD
" "	FORAGE HARVESTER	<75	POOR TO FAIR	GOOD
" (MATURE)	COMBINE, STRAW REMOVED	<75	POOR	FAIR
FESCUE-CLOVER MEADOW	FORAGE HARVESTER	<75	POOR TO FAIR	GOOD
SUDAN GRASS (12" HIGH)	MOWER	<75	POOR	FAIR
" "	FLAIL HARVESTER	<75	POOR	GOOD
RYE (FULL GROWTH)	MOW, RAKE, BALE	<75	POOR	GOOD
" "	FORAGE HARVESTER	<75	POOR TO FAIR	GOOD
" (MATURE)	COMBINE, STRAW REMOVED	<75	POOR	FAIR
WHEAT (MATURE)	" " "	<75	POOR	FAIR
CORN (FULL GROWTH)	FORAGE HARVESTER	<75	POOR	FAIR
MULCH (5 TONS WHEAT STRAW/ACRE)	SIDE-DELIVERY RAKE	75-95	POOR	FAIR
" (5 TONS BERMUDAGRASS HAY/ACRE)	RAKE AND BALE	<75	POOR	GOOD
RATING OF REMOVAL: GOOD 5 ACRES/H, FAIR-1 to 5 ACRES/H, POOR 1 ACRE/H RATING OF DISPOSAL (ADDITIONAL LOADING AND HAULING): GOOD-MINIMAL, FAIR-CONSIDERABLE, POOR-VERY GREAT				

Table 5.11 Removal of vegetation and crops

ADVANTAGES:

- REDUCE THE AREA CONTAMINATION ABOUT 50%
- NO DISTURBANCE OF SOIL LAYER AND PRODUCTIVITY

DISADVANTAGES:

- DEPENDENT ON
 - , KIND AND DENSITY OF VEGETAL COVER
 - , CROPS GROWING STATE
 - , DRY OR WET DEPOSITION
 - , SEASON FOR NATURAL PERENNIAL WOODS
 - , PERIOD OF TIME BETWEEN CONTAMINATION, CUTTING AND TAKING AWAY
- MUST START AS SOON AS POSSIBLE
- REQUIRES CONSIDERABLE TIME
- INVOLVES PROBLEMS OF WASTES TREATMENT AND DISPOSAL
- ENVIRONMENTAL IMPACT
- DECADES WILL BE REQUIRED FOR REVEGETATION IN NATURAL ECOSYSTEMS.

FURTHER KNOWLEDGE NEEDED

- EFFICIENCY, PRACTICABILITY AND TIME REQUIRED FOR EACH METHOD OF VEGETATION REMOVAL, ECOSYSTEM INVOLVED, TYPE OF CROPS, AND CONTAMINATION LEVEL
- NORMS AND METHODS FOR PERSONNEL PROTECTION DURING THE EXECUTION OF EACH METHOD
- TREATMENT AND DISPOSAL FOR REMOVED VEGETATION
- ESTABLISHMENT OF REFERENCE LEVELS FOR APPLICATION OF THIS COONTERMEASURES IN DIFFERENT ECOSYSTEMS

5.3.2.2 CHEMICAL TREATMENT OF SOILS

- Leaching

The leaching of contaminants by irrigation is not expensive and it reduces the wind-blown material.

Depending on the type of soil and its profile, the depth mobility of the contaminants is generally low. The soil productivity could be affected (essential nutrient elements could be moved with the contaminants), and the addition of fertilizers would be necessary.

As in the ploughing, the radionuclides could still be available to plants.

Table 5.12 shows the advantages and disadvantages of the irrigation and leaching.

Table 5.12 Irrigation and leaching

ADVANTAGES

- NOT EXPENSIVE
- NO WIND-BLOWN MATERIAL NOR MECHANICAL RESUSPENSION

DISADVANTAGES

- LOW DEPTH MOBILITY OF CONTAMINANTS (DEPENDANCE ON SOIL PROFILE)
- DECREASE SOIL PRODUCTIVITY (ESSENTIAL NUTRIENT ELEMENTS MOVE WITH CONTAMINANTS)
- ADDITION OF FERTILIZERS IS NEEDED
- RISK OF SURFACE AND SUBSURFACE WATERS CONTAMINATION
- LARGE AMOUNT OF WATER NEEDED

- Fixation of contaminants with road oil, asphalt, foam...

Their application stops the spreading and resuspension of contaminants, and increases the effectiveness of latter operations which involve the removal of the contaminated surface. These methods could be used as an initial step for further decontamination operations.

When the fixatives are applied, no waste is generated, and their use may be less expensive than other treatments. It's not permanent, but could last many years.

Their main disadvantages are: The land is almost useless during the period which it is covered, some drainage is needed and water would not penetrate.

Table 5.13 shows the advantages and disadvantages and further knowledge needed in relation with the application of fixatives to soils.

- Applications of lime, fertilizers and others amendments

A reduction of radionuclides uptake is achieved by providing cations competition, precipitation or high cation exchange capacity products.

Their main advantages are: Can be more easily applied than most other treatments for contaminated land, allow a selective uptake reduction of radionuclides and correct the soils fertility and acidity.

None of the soil amendments are highly effective. Their use depends on the type of soils, the kind of radioactive contamination and on the availability of large amount of the amendment to be applied.

An evaluation of the chemicals used is presented in table 5.14.

A comparison of soil amendments for reducing Sr-90 uptake from contaminated soils is presented in table 5.15.

Table 5.16 summarizes the advantages, the disadvantages and the further knowledge needed in relation of the soil amendments application.

Table 5.13 Application of fixatives to soils

ADVANTAGES:

- IS A GOOD TEMPORARY COUNTERMEASURE TO:
 - . REACH MAXIMUM REDUCTION OF RADIONUCLIDES RESUSPENSION BY WIND OR ANY PHYSICAL DISTURBANCE
 - . GAIN OF TIME UNTIL DECISIONS ARE MADE ON:
 - PLANTS REMOVAL
 - SURFACE SOIL REMOVAL
 - TREATMENT OF VEGETATION AND SOIL
 - WASTES DISPOSAL (VEGETATION AND SOIL)
- COULD LAST MANY YEARS, NOT PERMANENT
- MOST PLANTS CAN REMAIN IN SITU
- ALMOST 85% DECONTAMINATION BY TAKING OUT THE FIXATIVES (FOAM)
- MAY BE LESS EXPENSIVE THAN OTHER METHODS

DISADVANTAGES:

- LAND IS ALMOST USELESS DURING THE COVERED PERIOD
- VERY RESTRICTIVE FOR ANIMAL LIFE
- REQUIRES A CONSTANT VIGILANCE OF CONTAMINANTS UNDER COVER
- DRAINAGE NEEDED, WATER WOULD NOT PENETRATE
- RAINFALL RUNOFF MAY BE INCREASED
- MOST OF FIXATIVES COULD HAVE SERIOUS ENVIRONMENTAL EFFECTS
- MOST ARE APPLICABLE TO SMALL AREAS ONLY

FUTHER KNOWLEDGE NEEDED:

- ESTIMATION OF ECOLOGICAL IMPACT ALONG THE TIME FROM TEST MADE
- RELATION BETWEEN APPLICATION OF FIXATIVES AND LOSS OF SOIL PRODUCTIVITY FOR DIFFERENT CROPS

Table 5.14 Evaluation of the chemicals use and restrictions in the scenarios

SCENARIO	METHOD	EFFICIENCY	COST	PRACTICABILITY
Rocky Flats Plant	Polyurethane foam	Depends on the surf.: In a 0.6Ci/m ² zone the effic. is 85% in a single application of 5 cm deep	5 cm thick coating foam → 36 pound per 10 m ² for flat surface. Cost material: \$24 per 10m ² (1973)	Durability is excellent. Plants covered: - totally → killed. - partially → survived
Nevada	Oiling	70% of surface contamination reduction		Good if associated with plowing
Chernobyl	Lime, zeolite and inorganic fertilizer			
Windscale	Milk consumption restriction when > 3700 Bq/l	Good		Good

Table 5.15 Comparison of soil amendments for reducing Sr-90 uptake from contaminated soils

METHOD	REDUCTION IN ⁹⁰ Sr UPTAKE	EFFORT REQUIRED	EFFECT ON SOIL PRODUCTIVITY
LIME APPLICATIONS TONS 2 TO 10/ACRE	50-95%	GOOD	GOOD
NITROGEN FERTILLIZERS, 100# N/ACRE	<75%	GOOD	GOOD
PHOSPHATE FERTILLIZERS, 100# P/ACRE	<75%	GOOD	GOOD
POTASSIUM FERTILLIZERS, 500# K/ACRE	<75%	GOOD	GOOD
ORGANIC COMPOST, 5 TO 20 TONS/ACRE	<75%	FAIR	GOOD
CLAY MINERALS, 5 TO 20 TONS/ACRE	<75%	FAIR	GOOD TO FAIR
AMMONIUM OR POTASIUUM PHOSPHATE, 2 TO 5 TONS/ACRE	75-95%	FAIR	FAIR TO POOR
RATING OF EFFORT REQUIRED: GOOD-NOT SIGNIFICANTLY MORE THAN NORMAL FIELD PRACTICES FAIR-EXTRA EQUIPMENT, MATERIALS OR LABOR REQUIRED POOR-VERY GREAT REQUIREMENT OF EQUIPMENT, MATERIALS, OR LABOR RATING OF EFFECT ON SOIL PRODUCTIVITY: GOOD-INCREASE OR DOES NOT CHANGE PRODUCTIVITY FAIR-REDUCES PRODUCTIVITY < 20 PERCENT POOR-REDUCES PRODUCTIVITY > 20 PERCENT			

Table 5.16 Soil amendments (lime, fertilizers)

<u>ADVANTAGES</u>
- COULD BE MORE EASILY APPLIED THAN OTHER TREATMENTS - SELECTIVE UPTAKE REDUCTION OF RADIONUCLIDES - CORRECTION OF SOILS FERTILITY AND ACIDITY PROBLEMS
<u>DISADVANTAGES</u>
- NONE OF THE SOIL AMENDMENTS ARE HIGHLY EFFECTIVE - APPLICATION DEPENDANCE ON: . TYPE OF SOILS . . CHEMICAL STATE OF CONTAMINANTS RADIONUCLIDES . AVAILABILITY IN PLACE OF AMENDMENT
<u>FURTHER KNOWLEDGE NEEDED</u>
- EFFECTS OF SOIL AMENDMENTS ON REDUCTION OF SOME RADIONUCLIDES UPTAKE AND REDUCTION OF SOIL PRODUCTIVITY FOR DIFFERENT TYPES OF SOIL

5.3.2.3 MECHANICAL TREATMENTS OF SOIL

- Removal of soil

The complete removal of a soil layer is nearly 100% effective, not excessively costly and can be done rapidly.

The soil removal involves the use of special machinery, creates a large amount of waste, produces resuspension, and has a very important environmental impact which could be corrected by the addition of another fresh layer of soil or fertilizers.

The revegetation could be extremely difficult, but would become easier if, during the soil removal, a part of the vegetation remains in place.

Table 5.17 shows an evaluation of efficiency, cost and practicability of this countermeasure per scenario where it was applied.

Table 5.18 shows an evaluation of soil decontamination methods at Nevada Test Site.

Table 5.19 presents the more efficient decontamination methods of soil removing in normal weather conditions.

The advantages, disadvantages and further knowledge needed in relation of soil removal are summarized in table 5.20 when the removal is complete and in table 5.21 when some vegetation is left on the soil surface.

Table 5.17 Evaluation of the removal of soils in the scenarios

SCENARIO	METHOD	EFFICIENCY	COST	PRACTICABILITY
Los Alamos	Backhoe, crawler tractor, bulldozer front-end loader	Fraction remaining (FR): 10^{-2}	Excavation: \$50/m ³ (1976) \$769000 (1975-76) \$1037800 (1981-83)	In 1 year 15000 m ³ of tuff were broken-up to 1 m depth
Bikini	⇒Remove debris ⇒Radioactive decay			
Enewetak	Top 20 cm (>2220 Bq/kg)		84000m ³ ($\approx 4.7 \cdot 10^{11}$ Bq) Total cost: \$100 million	
Nevada	⇒Vacuum ⇒Front end loader ⇒Shovel + bags ⇒Debris and soil removal	⇒Successful under difficult environmental conditions FR: 10^{-3} ⇒FR: 10^{-2} ⇒FR: 10^{-1} to 10^{-2}	⇒Cost reduction due to volume reduction ⇒Total cost(1978-1982) \$1.28million/850 tons	⇒It is possible to remove less than 5 cm of soil per pass. No disturbance of the vegetation root system. High decont. factor in fine grain terrain. ⇒Rapid and good decont.factor. ⇒Slow, but can be used in locations inaccessible to heavy equipment.

Table 5.17 Evaluation of the removal of soils in the scenarios
(Continuation)

SCENARIO	METHOD	EFFICIENCY	COST	PRACTICABILITY
Rocky Flats Plant	⇒Hand excavation	⇒Save and good	⇒Costly 35 boxes(4'x4'x7') \$1057/box	⇒Good but time consuming
	⇒Mechanized equip. (front-end-loader and/or road maintainer dozer)		⇒1488 boxes(2'x4'x7') \$ 283/box	⇒Good unless where was no possible to use
Palomares	10 cm removed with 3 road graders 2 bulldozers 2 front-end-loader	Good	831 m ³ into 5300 barrels	Good Problems in an isolated hilly area (→ hand removal)
Thule	Ice removal	Very efficient 98%	1200 m ³ removed by 700 persons during ≈ 7.5 months	Good
Chernobyl	Top 5-10 cm Scraping with bulldozers	Contamination decreased by a factor of 20 to 30		Problem of vision with the remotely controlled machinery

Table 5.18 Evaluation of soil decontamination methods at Nevada Test Site

TERRAIN	DECONTAMINATION METHOD	ESTIMATED FRACTION REMAINING(FR)	METHOD OF EVALUATION
FLAT COMPACT DIRT, FINE GRAIN	FRONT END LOADER	10^{-2}	POSITIVE: RAPID, GOOD DECON. FACTOR NEGATIVE: INEXPERIENCED DRIVER CAN RECONTAMINATE CLEAN AREAS
	VACUUM	10^{-3}	POSITIVE: HIGH DECON FACTOR, GOOD WHERE SUBSURFACE BOULDERS ARE PRESENT, NO EXPERIENCED OPERATORS REQUIRED NEGATIVE: SLOW COMPARED TO FEL
	SHOVEL & BAGS	10^{-2}	POSITIVE: GOOD IN LOCATION INACCESSIBLE TO HEAVY EQUIPMENT NEGATIVE: SLOW CONSIDERABLE CARE MUST BE TAKEN NOT TO RECONTAMINATE CLEANED AREAS (EXPERIENCED PERSONNEL DESIRABLE)
FLAT GROUND, COARSE GRAIN OR GRAVEL-LIKE	FRONT END LOADER	10^{-1} TO 10^{-2}	POSITIVE: MOVES LARGE AMOUNTS OF DIRT NEGATIVE: INEXPERIENCED DRIVER CAN RECONTAMINATE CLEAN AREAS
	VACUUM	NOT ATTEMPTED	
	SHOVEL & BAGS	10^{-1}	POSITIVE: CAN BE USED IN LOCATIONS INACCESSIBLE TO HEAVY EQUIPMENT NEGATIVE: IMPRACTICAL IN LOCATIONS WHERE DEEP PENETRATION OF CONTAMINATION HAS OCCURRED
ROCKY CANYON (SPRING DRAINAGE BED)	VACUUM	NOT ATTEMPTED	POSITIVE: CAN BE USED IN MAXI COLLECTION AREAS INACCESSIBLE TO FEL BY ADDING EXTENSION HOSE NEGATIVE: MOST ROCKY AREAS INACCESSIBLE TO VACUUM EVEN WITH HOSE EXTENSION
	SHOVEL & BAGS	10^{-1} (FINE GRAIN SOIL)	POSITIVE: GOOD IN LOCATIONS WHERE CONTAMINATED MUD HAS COLLECTED NEGATIVE: USUALLY TOO MANY ROCKS
	FLUSHING WITH WATER HOSE OR SCRUBBING WITH BROOMS/BRUSHES	10^{-3} (ROCKY SURFACES) 10^{-1} (CRACKS)	POSITIVE: GOOD IN AREAS WHERE CONTAMINATION IS HIDDEN UNDER ROCKS OR EXPOSED ON ROCKY SURFACES NEGATIVE: CARE MUST BE TAKEN SO CONTAMINATION IS NOT SPREAD FURTHER; E.G. CONSTRUCTING A DAM

Table 5.19 More efficient decontamination methods of soil removing in warm conditions

SURFACE CONDITIONS	METHOD	EFFICIENCY (%)	EFFORT REQUIRED FOR DISPOSAL
BLUEGRASS SOD	SOD CUTTER 12" WIDE	>95	CONSIDERABLE
FESCUE-CLOVER 12" HIGH	MOTOR GRADER	>95	GREAT
PLOWED	BULLDOZER	>95	GREAT
PLOWED	PAN-TYPE SCRAPER	>95	CONSIDERABLE
SEEDBED	MOTOR GRADER	>95	GREAT
SEEDBED	BULLDOZER	>95	GREAT
SEEDBED	PAN-TYPE SCRAPER	>95	CONSIDERABLE
* THE EFFORT REQUIRED FOR REMOVAL IS, IN ALL CASES, EVALUATED AS <1 ACRE PER HOUR			

Table 5.20 Complete removal of soil surface

ADVANTAGES:

- DECONTAMINATION COULD BE ALMOST 100%
- CHEAPER THAN LEAVING SOME SHRUBS
- CAN BE DONE RAPIDLY

DISADVANTAGES:

- GROUND COVER WILL BE REDUCED AND PRIMARY PRODUCTIVITY WILL BE DECREASED IN SOME DEGREE
- REVEGETATION WOULD BE EXTREMELY DIFFICULT UNLESS MICRO-DEPRESSIONS FOR ARTIFICIAL REPLANTING BE MADE ON PREVIOUS SITES OF SHRUBS
- FERTILIZERS, ORGANIC MATTER OR FRESH SOIL MIGHT BE REQUIRED
- NATIVE ANIMALS WOULD BE LOST ALTHOUGH THEY WOULD BE REPLACED WITH THE TIME FROM ADJOINING AREAS
- DUST-BOWL CONDITION WOULD EXIST UNLESS SOIL IS STABILIZED OR VEGETATION AGAIN GROWS
- BIG AMOUNT OF WASTES ARE PRODUCED
- THE TYPE OF TERRAIN IMPOSE CONDITIONS TO THE MECHANIZED EQUIPMENT TO BE USED

FUTHER KNOWLEDGE NEEDED:

- DEVELOPMENT OF EQUIPMENT AND TECHNIQUES FOR CONSTRUCTION OF MICROCATCHMENT DEPRESSIONS TO COLLECT AND CONCENTRATE RUN OFF
- DEVELOPMENT OF SEEDING AND TRANSPLANTING TECHNIQUES FOR REVEGETATION IN THE CATCHMENT BASINS
- ELABORATION OF A LIST OF PLANTS AND CROPS WHICH COULD GROW IN DISTURBED SOIL AREAS
- EVALUATION OF SOIL PRODUCTIVITY REDUCTION AND RECOVERY FOR SPECIAL PLANTS AND CROPS
- IMPROVE TECHNIQUES FOR WASTE REDUCTION AND DISPOSAL

Table 5.21 Removal of surface soil leaving some vegetation

ADVANTAGES:

- DECADES WILL NOT BE REQUIRED FOR REVEGETATION OF NATURAL ENVIRONMENTS
- REMAINING VEGETATION PROVIDES SEED FOR CONTINUED REVEGETATION AND PRESERVE FERTILE SHRUB SITE FOR NEW ARTIFICIAL SEEDLING
- PROTECTION AND FEED ARE ASSURED FOR NATIVE ANIMALS
- COST OF REVEGETATION IS LARGELY REDUCED OR AVOID
- DUST RESUSPENSION IS REDUCED, ESPECIALLY IF STABILIZERS ARE USED
- THE AREA IS REASONABLY ENVIRONMENTALLY AND ESTHETICALLY ACCEPTABLE

DISADVANTAGES:

- DECONTAMINATION MAY NOT BE 100% EFFECTIVE
- GROUND COVER WOULD BE DIMINISHED WITH SOME REDUCTION OF PRIMARY PRODUCTIVITY
- EROSION FROM RAINFALL COULD BE ACCELERATED UNLESS A STABILIZATION OF SOIL BE MADE
- INVOLVE PROBLEMS RELATED WITH AMOUNT OF WASTES
- CLEAN UP PROCESS IS MORE COSTLY THAN COMPLETE SURFACE SOIL REMOVAL
- ENORMOUS DUST RESUSPENSION DURING LOADING AND TRANSFERING OPERATIONS MUST BE SOLVED
- THE TYPE OF TERRAIN IMPOSES CONDITIONS TO THE MECHANIZED EQUIPMENT USED
- INEXPERIENCE OPERATING PERSONNEL COULD RECONTAMINATE CLEAN AREAS

FURTHER KNOWLEDGE NEEDED

- DEVELOPMENT OF EQUIPMENT FOR REMOVING SOIL AROUND SHRUBS WITHOUT DESTROYING THEM
- IDENTIFICATION OF PLANTS DIFFICULT TO CLEAN AROUND
- METHODS TO REDUCE OR SOLVE IN THE SMALLER TIME, PROBLEMS OF TAKING OUT THE FERTILE LAYER OF SOIL
- PILOT TEST TO ASCERTAIN WIND AND RAIN EROSION EFFECTS AFTER SUCH COUNTERMEASURES
- IMPROVE VACUUM EQUIPMENT FOR APPLICATION AT LARGE AREAS
- IMPROVE TREATMENT FOR WASTE REDUCTION AND DISPOSAL

- Deep placement of contamination

By turning the soil the contaminants are placed deeper than the root depth layer to avoid their availability by the crops. The exposure dose rate is significantly reduced.

During the operations, the resuspension could be minimum.

Disturbance of soil layers and loss of fertility is produced.

Revegetation is needed and would be sparse for decades.

Recently, some activities are developed to modify a conventional plough so that it could skim-off the topmost 5 cm soil and bury it beneath some 50 cm soil without inverting the 5-50 cm horizon, in order to avoid loss of fertility.

Table 5.22 shows the advantages, disadvantages and further knowledge needed of this countermeasure.

- Normal and deep ploughing

The radionuclides availability to plants and animals depends on the dilution of contaminants achieved and on the root depth of crops.

The radiation levels of exposure and the resuspension factors are reduced.

A highly effective reduction in radionuclides uptake can be achieved by simultaneous use of some chemicals which stop the root growth.

The normal ploughing can be made by conventional farm machinery and procedures, and it is one of the cheapest effective methods for soil treatment.

The radionuclides could still be available to plants although they were greatly diluted.

The efficiency, cost and practicability of ploughing is presented in table 5.23.

Table 5.24 summarized the advantages, the disadvantages and the further knowledge needed in relation of this countermeasure.

As a summary of the possible remedial actions to be applied for direct treatment of soils, in table 5.25 the representative cost and productivity data for agricultural fields decontamination operations are shown.

Table 5.22 Deep placement of contaminated surface soil

ADVANTAGES:

- ALMOST 100% DECONTAMINATION OF SURFACE SOIL
- RADIONUCLIDES ARE NOT AVAILABLE TO CROPS
- NO RESUSPENSION PROBLEMS OF RADIONUCLIDES
- IMPORTANT REDUCTION OF RADIATION EXPOSURE RATE LEVELS

DISADVANTAGES:

- DISTURBANCE OF SOIL LAYERS AND FERTILITY
- IN NATURAL ECOSYSTEMS REVEGETATION WILL BE NEEDED AND WOULD BE SPARSE FOR DECADES
- COMPOSITION OF UNDERNEATH SOIL LAYERS COULD BE MADE NOT SUITABLE THIS COUNTERMEASURE

FURTHER KNOWLEDGE NEEDED:

- DEVELOPMENT OF SUITABLE EQUIPMENT FOR APPLICATION IN LARGE AREAS
- ELABORATION OF A LIST OF PLANTS DEPTH ROOTS TO TAKE INTO CONSIDERATION THE OPTIMA DEPTH PLACEMENT
- LIST OF ADDITIVES TO CORRECT THE LOSS OF SOIL FERTILITY BY TURNING DOWN THE SURFACE SOIL IN DIFFERENT TYPES OF SOIL

Table 5.23 Evaluation of ploughing in the scenarios

SCENARIO	METHOD	EFFICIENCY	COST	PRACTICABILITY
Enewetak	Front-end-loader (50 cm depth)	Relatively effective mixing surface down 50 cm; ≈ tenfold decontami- nation factor	Plowing speed: 67 m/min	Satisfactory after devegetation. Deep disking following the plowing would have mixed the soil more uniformly as is the case in usual agricultural practice.
Nevada	⇒Plowing ⇒Oiling + scraping ⇒Leaching +scraping ⇒Disking ⇒Water leaching ⇒Scraping ⇒Oiling	⇒97.9% of surface ⇒95.6% contamination ⇒92.7% reduction ⇒89.2% ⇒87.4% ⇒86.0% ⇒69.4%	⇒) Cheapest ⇒)	⇒Very good
Palomares	⇒Front-end-loader (30 cm depth) Remove debris, raking, mixing and watering	⇒Dilution factor: from .01 cm to 45 cm is $4.5 \cdot 10^5$		Unless in a hill (made by hand digging) Good

Table 5.24 Normal and deep ploughing

ADVANTAGES:

- REDUCE THE RADIATION LEVELS OF EXPOSURE
- REDUCE THE RESUSPENSION FACTORS OF RADIOACTIVITY
- REDUCE THE AVAILABILITY OF RADIONUCLIDES TO PLANTS IN FUNCTION OF ITS ROOTS DEPTH
- CAN BE MADE BY CONVENTIONAL FARM MACHINERY AND PROCEDURES
- IT IS ONE OF THE CHEAPEST AND MORE EFFECTIVE METHODS FOR SOIL TREATMENT
- DISKING FOLLOWING PLOWING DECREASE THE HETEROGENEITY OF THE RADIONUCLIDES MIXING

DISADVANTAGES:

- RADIONUCLIDES ARE STILL AVAILABLE TO PLANTS ALTHOUGH GREATLY DILUTED
- NOT USEFUL FOR HIGH LEVELS OF RADIOACTIVE CONTAMINATION
- POOR CROPS AFTER DEEP PLOWING WILL BE PRODUCED IN MANY SOILS
- FERTILIZERS AND LIME ADDITION TO THE NEW TOPSOIL WILL BE NECESSARY AFTER DEEP PLOWING
- IMPERVIOUS CALICHE LAYERS AND ROCKS COULD MAKE IT DIFFICULT

FURTHER KNOWLEDGE NEEDED:

- ELABORATION OF A LIST OF PLANTS DEPTH ROOTS TO TAKE INTO CONSIDERATION THEIR OPTIMA DEPTH TO PLOW
- ADDITIVES TO CORRECT THE LOST OF SOIL FERTILITY BY DEEP PLOWING IN DIFFERENT TYPES OF SOIL

Table 5.25 Summary of representative cost and productivity data for agricultural fields decontamination operations

OPERATION	RATE (m^2/HR)	COST (1982 \$/ m^2)			
		TOTAL	LABOR	EQUIPMENT	MATERIAL
WATER	2,149	0.0219	0.0092	0.0127	
FIXATIVE, COHEREX	2,922	0.2061	0.0068	0.0094	0.19
LEACH, FeCl_3	1,814	0.052	0.0109	0.0151	0.026
SCRAPE	875	0.31	0.13	0.18	
PLOW 10-12 INCHES	8,500	0.004	0.001	0.002	
DEEP PLOW	5,000	0.06	0.005	0.055	
CLEAR	543	0.026	0.009	0.017	
COVER WITH SOIL	549	0.371	0.106	0.265	

ACTUAL PRICES FOR SOME OPERATIONS IN PALOMARES AREA ARE:

- PLOWING: 12000 PTS/HA (\$100/HA)
- REMOVAL OF TOP SOIL: 3000 TO 4000 PTS/H (\$25 TO 33/H)
- TRUCKS MOVING 4 TIMES THE DISTANCE OF 25 TO 30 KM IN 8 HOURS OF WORK: 2500 PTS/H (\$21/H)
- THE AVERAGE PRICE FOR REMOVAL, LOADING AND TRANSPORTING SOIL: 900 PTS/ m^3 (\$7.5 m^3)

5.3.2.4 ALTERNATIVES TO TREAT CONTAMINATED SOIL

Since the major hazard from farmland contamination arises from the entry of radionuclides into human food and because of the great effort required for effective land decontamination, some alternatives to soil treatment have been suggested.

These alternatives are summarized in table 5.26.

Table 5.26 Alternatives for soil treatment

- TO GROW CROPS WITH SMALL RADIONUCLIDES CONTRIBUTION TO HUMAN DIET
- TO USE CONTAMINATED PASTURES FOR BEEF OR MUTTON INSTEAD OF DAIRY PRODUCTION
- TO REMOVE RADIONUCLIDES FROM MILK AND OTHER PRODUCTS BY SPECIAL TREATMENTS IN PROCESSING
- TO USE THE PRODUCTS WITH LOW RADIONUCLIDES CONCENTRATION FROM CROPS PROCESSING
- TO GROW CROPS FOR ANIMAL FEEDING AND NOT FOR DIRECT HUMAN COMSUMPTION
- TO GROW CROPS FOR INDUSTRIAL USES

- Choice of crops

Some crops contribute little or nothing to the radioactive contents of the human diet, even if they were grown on highly contaminated soils: Fiber crops, such as cotton and flax, are obvious examples. Sugar and oil crops would have most of the radioactive materials in the parts removed from the refined products. However, in the case of byproducts, such as cottonseed meal or sugarbeet pulp, which are used to feed animals, the indirect contribution of radionuclides to the human diet would have to be considered.

Since corn has one of the lowest mineral contents of any grain, its content of radionuclides, such as strontium, is very low. Other essential food crops, especially those with an important contribution to the minerals of the diet, would have to be grown on land with small radionuclides contamination. Such crops include fruits and vegetables.

When the contaminating material is strontium, the use of the land for beef, pork, mutton or poultry production would be advantageous, because meat and eggs have a small contribution to the radioactive strontium contents in the human diet. This may not be true when other radionuclides constitute the main hazard. For example, meat contributes, almost as much as milk, to the cesium-137 in the diet.

Although the alternatives to treating contaminated soil would be fully used, the land for nutritionally critical crops should be treated preferentially. Critical crops might vary, depending on the crops that are normally produced in areas which have resulted in the high contamination and the possibility of transporting substitutes of that crops from other areas. In subsequent years, more land could be treated for producing critical crops.

Anyway, as the Danish study (chapter 3.3) has shown, by choosing of some species of crops it would be possible to cultivate the same crops as before if the level of contamination is not as high as it might be in the far and maybe midle field.

- Food processing

Ion-exanchage treatment of milk could reduce its strontium-90 content perhaps more effectively than decontamination treatments or management of hay and pasture land. In full-scale test of ion-exchange treatment in a milk-processing plant, from 90% to 97% of the strontium-90 was removed from the milk. Similar treatment may be possible with vegetable and fruit juices and purées. However, some esential nutrients are also removed.

In many situations by a combination of treatement and alternatives, it might be possible to use very highly contaminated land for agricultural practices. In fact after the treating of soils (by the adecuate chemical and/or mechanical treatment) it seems eficacious control the radionuclides tranfer into the food chain by the using of one of the above alternatives related to the produced crops and food.

6. FURTHER RESEARCH NEEDS

With reference to the CEC Post-Chernobyl Action n° 6 "Countermeasures in the Agricultural Environment" and from information contained in the participants' contribution in this report compiled, the following summarized considerations could be pointed out about subjects which need further knowledge and will require some studies or research effort:

- The behaviour of soil and plant radionuclide uptake as a function of their physico-chemical state as released during accidents in pressure and boiling water reactors.
- The development of guides and tables on
 - . soil-crop transfer factors for the most important radionuclides, types of soil and climatological conditions. Chemical amendments to reduce the concentrations of contaminants and uptake by different crops;
 - . the impact of the removal of fertil soil layers on the productivity of different crops and procedures to recover the soil fertility for the usual or alternative crops;
 - . the type of plants and crops which could grow in disturbed soil areas;
 - . the root depth of crops and its influence when deep ploughing and deep placement of contaminants are carried out;
 - . the migration of surface contaminants in different types of soil and its influence on the contamination of underground waters and water stores.
- The study of resuspension of contaminated soil particles and their indirect quantitative impact on crops.
- The development of techniques and machinery for deep placement of contaminants.
- The improvement of vacuum equipment for surface soil removal over large areas.
- The development of techniques and machinery for the laying on of microcatchment depressions on soil and for revegetation in the catchment basins.
- The treatment for waste reduction and disposal of radioactive vegetation and soil removed.
- The alternatives to land recovery
 - . crop management;
 - . processing of food products (vegetables, fruits, etc.).
- The models and specific parameters to determine the impact of agricultural countermeasures over a period of time in the most typical environments of Europe.

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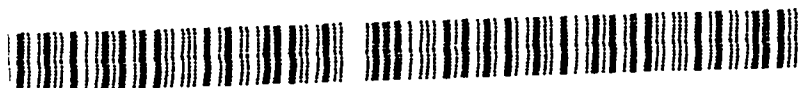
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