Review of Past Nuclear Accidents: Source Terms and Recorded Gamma-ray Spectra.

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Abstract:
Airborne gamma ray spectrometry using high volume scintillation detectors, optionally in conjunction with Ge detectors, has potential for making rapid environmental measurements in response to nuclear accidents. A literature search on past nuclear accidents has been conducted to define the source terms which have been experienced so far. Selected gamma ray spectra recorded after past accidents have also been collated to examine the complexity of observed behaviour.

SUMMARY

Airborne Gamma Spectrometry (AGS) has developed considerably since the Chernobyl accident, and is more widely recognised as having an important contribution to make to nuclear emergency response. The abilities to survey large areas rapidly with high sampling densities, and to produce radiometric maps on emergency timescales have already been demonstrated. However the ability of AGS systems to resolve a complex gamma ray spectrum, particularly in the early phases of a reactor accident is less well defined. This report reviews past experience of nuclear accidents, and the gamma ray spectra recorded immediately afterwards, particularly from the air, in preparation for an experimental and theoretical study of the AGS response to a fission product source.

A literature survey of past nuclear accidents has been conducted, and used to prepare a list of accidents with off-site consequences. The accidents reviewed have occurred at a mean rate of 3 per year, since 1945. Accidents involving core damage have occurred every 2 to 3 years, with destruction of the core every 6 to 7 years. About 20% of the accidents resulted in some off-site contamination, with a further 10% resulting in contamination of the building. There were four accidents resulting in the release of over 10^{15} Bq, and one in which over 10^{18} Bq was released. Source terms for selected accidents; an explosion at a reprocessing plant in Chelyabinsk, the fire at Windscale, the crash of the Soviet satellite Cosmos-954, an explosion on a Soviet submarine during refuelling and the Chernobyl accident; are discussed.

Details of some of the gamma-ray surveys conducted after nuclear accidents are given, with particular emphasis on airborne surveys. A selection of γ-ray spectra from some of these surveys is presented. The influence of accident conditions, and post accident delays, on source term and spectral simplification has been noted. This information will be used, together with the results of experimental work in progress to evaluate data analysis strategies for high volume sodium iodide (NaI) spectrometers in the early stages of emergency response, and to examine the need for high resolution detectors.

ACKNOWLEDGEMENTS

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CONTENTS

1. INTRODUCTION

This report sets out interim findings of a study carried out for the Department of the Environment (DOE) to investigate the capability of Airborne Gamma Spectrometry (AGS) systems in measuring the quantities of short-lived fission products arising from nuclear and other radiological accidents. The aspects of the study that are reported here are the numbers and kinds of accidents that have occurred in the past, and the nature of the contamination, and hence measurement spectra, that they have given rise to. This information will in turn be used in subsequent parts of the study, along with other data, to help evaluate the capability of AGS systems in measuring post-accident contamination levels.

The report presents a brief introduction to the fission process and products, followed by identification of source terms from past nuclear accidents. Factors which influence the release of radionuclides are noted, and examples of the types of γ-ray spectra observed are shown.

2. BACKGROUND

2.1 Nuclear Fission

Nuclear fission is the process whereby heavy nuclei, typically actinides, split into two lighter nuclei, with an accompanying release of energy. For several nuclei fission follows the capture of a thermal neutron; such nuclei are said to be fissile. Nuclear reactors operate on a chain reaction in which a fissile nucleus (usually ^{235}U) absorbs a thermal neutron (ie neutrons which are in thermal equilibrium with the reactor, with energies of about 0.03eV) which induces fission. The fission releases energy and a number of fast neutrons, these neutrons are then moderated, slowing them down to thermal energies. If, on average, one of these neutrons is absorbed by another fissile nucleus resulting in fission then a self-sustaining chain reaction occurs, and the reactor is said to be critical. If, on average, more than one neutron is absorbed by a fissile nucleus then the reactor is said to be super-critical, if less than one neutron is absorbed the reactor is sub-critical.

Following fission a broad range of products is formed with masses mostly in the range from 60-170 amu; the yield of any one isotope being at most a few percent. Most of the nuclei formed are unstable, having an excess of neutrons, and decay by β-emission (with half lives ranging from milliseconds to several years), or very occasionally by neutron emission (with half-lives of less than a few seconds). Fission of ^{235}U induced by thermal neutrons forms about 250 different isotopes directly with independent yields in excess of 0.01%. Other isotopes are formed by the decay of these, usually by β-decay which results in isobaric decay chains where all the isotopes have the same mass. A typical β-decay chain is the decay of ^{131}In through several other isotopes, some of which are also formed directly in the fission process, to eventually form ^{131}Xe which is stable:

Fission yield(%):	0.034	0.034	1.38	0.15	0.0019	2.8×10^{-7}
	^{131}In	$\rightarrow$ ^{131}Sn	$\rightarrow$ ^{131}Sb	$\rightarrow$ ^{131}Te	$\rightarrow$ ^{131}I	$\rightarrow$ ^{131}Xe
Half life:	270ms	39.0s	23.0m	25.0m	8.4d	

This decay chain is significant because the radioactive isotopes of iodine are radiologically important, since if released into the environment they collect in the thyroid, resulting in an increased risk from thyroid cancers. Although the independent fission yield of ^{131}I is very low, substantial quantities will be present in a reactor because of the higher yields for parent isotopes, 1.6% of fissions will produce ^{131}I.

Figure 2.1 shows the mass distribution of fission fragments following thermal neutron induced fission of ^{235}U. The longer lived isotopes in the decay chains that are strongly populated (masses 88-103 and 132-144 have yields in excess of 3%) will be fairly common in the reactor, even if their independent fission yields are quite low. There are 187 isotopes formed with cumulative yields in excess of 1%, of which 82 have yields in excess of 5%. Of these 34 isotopes are stable, 8 have half lives in excess of 1000 years, 10 have half lives between 1 month and 1000 years, 11 have half lives between 1 day and 1 month, and 19 have half lives between 1 hour and 1 day, the remainder have half lives of less than 1 hour. Thus, despite the large number of isotopes produced by nuclear fission, relatively few will present any major

long term radiological hazard.

The radioisotope inventory of a nuclear reactor consists of a complex cocktail of fuel material, fission products, decay products and activation products caused by the capture of neutrons by other material in the reactor (fuel cladding, moderators, structural materials etc.). The composition of this inventory will change over time as fission products are produced, and then decay or absorb neutrons. Radionuclides with a short half-life will rapidly reach an equilibrium in the reactor, radionuclides with longer half-lives will continue to build up during reactor operation.

In the event of an accident the composition of the radionuclides released will depend upon the current radioisotope inventory of the reactor and the type and severity of the accident. These radionuclides may then be dispersed by atmospheric conditions and be deposited downwind of the accident. The distribution and radioisotope inventory of the resulting fallout will depend upon weather patterns and the time since the accident. Over time the radioisotope inventory of the fallout will change as a result of the nuclear decay of the radioisotopes, and within any part of the environment by various other processes that remove isotopes (for example being washed out by rain).

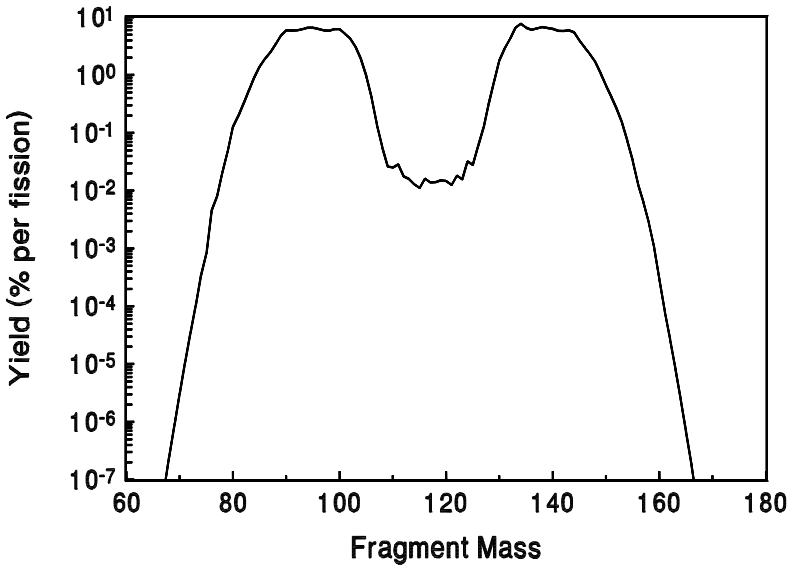

Figure 2.1: The mass distribution of fission fragments following fission of ^{235}U induced by thermal neutrons.

3. SOURCE TERMS

There are many potential accident types which might involve release of radioactive or nuclear material to the environment. The source terms for such events in this study are taken to be a description of the quantities and types of radionuclides, together with their physical/chemical form and mode of release.

3.1 Types of accident

There are many possible causes of nuclear accidents, including design faults, operator error, inappropriate operating procedures or damage to equipment. Some of the most common causes of damage to a reactor are loss of coolant, or reduction in coolant flow, and sudden changes in activity. Often, particularly in severe accidents, more than one cause contributed to the accident. Some of these causes are detailed in Appendix A.

Source terms calculated by Westinghouse for PWR reactors using the WASH1400 code (ref: USNRC 1975), and used in risk assessment for the Sizewell B PWR, are divided into four broad categories:
Release A: accidents of the severest type, the containment fails at the moment when the core becomes completely molten. A substantial amount of highly active radionuclides will then be released into the environment.
Release B: accidents where containment fails some hours after the core melts, there will be a substantial release of radionuclides although radioactive decay would have reduced the activity, and aerosol sedimentation would result in some retention.
Release C: accidents in which containment fails below ground level, but there is no substantial leak above ground.
Release D: accidents in which the containment does not fail, but a small release of activity may result at the design-rate.

Among the other, non-reactor, accidents that have occurred there have been accidents at other parts of the nuclear fuel cycle, including explosions at reprocessing, waste storage or fuel fabrication plants as a result of chemical or nuclear processes. Other accidents have involved the break-up of radioactive sources, for example from hospital equipment or nuclear weapons, in some cases with fragments being dispersed among the general population. There have also been accidents involving the re-entry of satellites containing radioactive materials. The nature of the accident determines both the magnitude of the source term and its isotopic composition.

3.2 Overview of past accidents

A search of available literature produced information on approximately 150 nuclear accidents, of which 38 resulted in off-site releases. These are listed in table 3.1, with a brief description of the incident and the consequences. The other incidents identified in the current work are given in Appendix B. Of these many were relatively minor, resulting in the release of no radioactivity or even damage to the core, but could have had more serious consequences. Some of the other accidents resulted in serious damage to the core, yet released little or no

radioactivity. Figure 3.1 shows various classes of incident divided into five year periods, with the classes being: the total number of incidents, the number of major incidents (ie: those involving radioactive releases, core damage, death or serious injury), the number of incidents in which only the buildings were contaminated, the number of accidents involving a release off-site, and the number of accidents involving a major release of radioactive material (ie: greater than 10^{15} Bq). The number of major incidents includes the incidents in the other categories.

From figure 3.1 it can be seen that nuclear accidents have occurred at the rate of approximately 3 per year. The accidents resulted in core damage in 20 cases and the destruction of the reactor in a further 8. Historically there have been accidents that damage the core, on average, every 2 to 3 years, and accidents in which the core is destroyed every 6 to 7 years. Of the accidents listed 38 (about 20%) resulted in some radioactive material being released to the environment, of which 4 released in excess of 10^{15} Bq and one over 10^{18} Bq, and a further 25 (about 10%) resulted in contamination of the building. On average there have been accidents at nuclear facilities resulting in a release of radioactive material every 2 years, with a significant release (in excess of 10^{15} Bq) once every decade.

It can be seen from figure 3.1 that there was an initial rapid increase in the number of reported incidents, corresponding to the increasing number of nuclear facilities. The rate of accidents then levelled off in the sixties and early seventies and is now declining slightly. However, the number of accidents involving off-site releases has remained approximately constant over this period. It is to be hoped that increases in knowledge, experience and public awareness will result in a reduction in the rates of accidents in the future, however past experience indicates a continuing need to be prepared to deal with an off-site release of radioactive material.

The more severe accidents at Chelyabinsk, Windscale and Chernobyl, the explosion while refuelling a submarine at Chazhma Cove and the crash of the satellite Cosmos-954, will be discussed in greater detail in the following section.

Date	Facility	Description of incident	Refs.
1951	Detroit, USA research reactor	Fissionable material overheated. The air was contaminated with radioactive gases.	Medvedev 1989
29/9/57	Chelyabinsk (Kyshtym) reprocessing plant, Southern Urals, USSR	A chemical explosion in concrete tanks holding highly active wastes released $7x10^{16}$ Bq of fission product to the environment. See section 3.1.3 for further details.	see section 3.3
10/10/57	Windscale graphite pile, UK	A fire released 10^{15} Bq of radioactive material into the atmosphere. See section 3.1.3	see section 3.3
26/7/59	Sodium Reactor Experiment, Santa Susanna, California	Sealant leaked into the cooling system, coating the fuel elements, and hence reducing the effectiveness of the coolant. The fuel elements overheated, and 10, out of 43, fuel assemblies were badly damaged. Some radioactivity was released from the stack.	IAEA 1994 Bertini et al 1980 Thompson and Beckerley 1964 McDonald and DeVan 1960
3/4/60	Westinghouse Testing Reactor, Pennsylvania, USA	Coolant flow was blocked by a piece of cladding from a fuel element. One element melted, and some radioactivity was released into the atmosphere.	IAEA 1994 Bertini et. al.1980 Thompson and Beckerley 1964 Korsmeyer 1960
3/1/61	SL-1, Idaho, USA experimental power reactor	A power surge, following the manual removal of a control rod, resulted in a pressure wave which lifted the pressure vessel 3m. Three people were killed by radiation and flying debris. The pressure vessel was distorted by the blast, about 10% of the fission products escaped from the pressure vessel, 0.01% from building. A further 23 people received significant radiation doses during the clean-up.	IAEA 1994 Bertini et. al.1980 Stratton 1967 Thompson and Beckerley 1964 Cottrell 1961, 1962 Buchanan 1963a
25/1/61	Reprocessing plant, Idaho, USA	After a release of compressed gas enriched U solution entered the evaporator. The area was highly contaminated.	Puit 1996
1962	Mexico	Four members of one family (including a child) died after contact with a $2x10^{11}$ Bq ^{60}Co source.	Pasquier 1996
1966	Palomares, Spain	An American B-52 bomber exploded, dropping its bombs which broke on impact. Pu was spread over 255 hectares.	Brown et. al.1996
1967	Lake Karachai, Kyshtym, USSR	After 1956 waste from the Kyshtym plant was dumped in Lake Karachai. As a result of a drought the lake dried out, and approximately $22x10^{12}$Bq of ^{137}Cs and ^{90}Sr was distributed as dust.	Brown et. al.1996
7/11/67	SILOE, France research reactor	During overpower testing a fuel element partially melted. $2x10^{15}$ Bq of fission products were released to the reactor pool, $7x10^{13}$ Bq of noble gases were released through the stack	IAEA 1994
1968	Thule, Greenland	An American bomber dropped its bombs, which split open on impact.	Brown et. al.1996
19/11/71	Monticello waste storage facility, Minnesota, USA	A tank overflow released 200 000 litres of contaminated water into the Mississippi.	Medvedev 1989
7/1/74	Leningrad NPP, USSR	An explosion of a reinforced concrete holder for radioactive gases.	Medvedev 1989
6/2/74	Leningrad NPP, USSR	An intermediate loop ruptured due to the boiling of water. Highly radioactive water was discharged into the environment.	Medvedev 1989
10/75	Leningrad NPP, USSR	Partial destruction of the core (local melt). About $5.5x10^{16}$ Bq of radionuclides were released through the ventilation stack.	Medvedev 1989
10/5/77	Dounreay	An explosion in a water filled shaft containing assorted highly active material, including fuel elements, occurred as a result of sodium-water reactions. Approximately 50 hot particles were recovered from the foreshore, outside the perimeter fence.	

13/12/77	Millstone-1 NPP, Connecticut, USA	Two explosions of hydrogen gas occurred in the off-gas stack building. One man was injured, and a small amount of radioactive contamination occurred.	Bertini et. al.1980 Casto et. al.1978
1978	Sétif, Algeria	One person died, and 5 more seriously injured (including 2 children) after a $6x10^{11}$ Bq Ir source was lost.	Pasquier 1996
24/1/78	Cosmos 954 satellite, Canada	A Soviet satellite crashed over northern Canada, the nuclear core was spread over a large area. See section 3.1.3.	see section 3.3
17/10/78	Reprocessing plant, Idaho, USA	U accumulated in a column as a result of an undetected flow of material. The area was highly contaminated.	Puit 1996
28/3/79	Three Mile Island NPP, Pennsylvania, USA	A pump in the turbine building stopped, which caused pumps downstream and the turbines to stop. The reactor scrammed. As a result of valves incorrectly left closed no water was supplied to the steam generators, which increased the temperature and pressure in the primary system. The pressure release valve opened, but failed to close, and primary water began to fill the drain tank. A faulty meter caused the operator to shut off water flow to the primary system, allowing a bubble of steam and hydrogen to form uncovering the top of the core, which partially melted. About $6x10^{11}$ Bq of ^{131}I, $1.6x10^{15}$ Bq of ^{85}Kr and other noble gases were released into the atmosphere, and liquid radioactive wastes discharged into the Susquehanna River. Most of the release occurred while eliminating the hydrogen bubble.	Hull 1988, 1989 Bertini et. al.1980 Casto and Cottrell 1979 NSAC 1979
7/8/79	Irving fuel fabrication plant, Tennessee	A discharge of highly enriched uranium.	Medvedev 1989
6/1/81	Reprocessing plant, La Hague	A fire in a storage silo released ~$4x10^{11}$Bq into the atmosphere, which was deposited close to the accident, with surface contamination reaching $11MBqm^{-2}$. Five people received a significant external dose, and a further 35 slightly exposed.	Joly 1996
25/1/82	R.E.Ginna NPP, Rochester, New York	A broken tube in a steam generator resulted in the release of radioactive steam into the atmosphere.	Medvedev 1989
30/1/82	Ontario NPP, New York	A breakdown in the cooling system caused a leak of radioactive substances into the atmosphere.	Medvedev 1989
9/82	Chernobyl NPP, USSR	The central fuel assembly of No.1 reactor was destroyed due to operator error. Radioactive material was released into the local area.	Medvedev 1989
1983	Ciudad Juarez, Mexico	4000 people were exposed after the dispersion of 6000 ^{60}Co particles from a $2x10^{13}$ Bq source scrapped without precautions.	Pasquier 1996 Brown et. al.1996
1984	Casablanca	A family was irradiated by a 10^{12} Bq Ir source collected by the father.	Pasquier 1996
19/5/85	Indian Point NPP, New York	The failure of a valve led to the discharge of several hundred gallons of radioactive water, some of which entered the environment beyond the facility.	Medvedev 1989
10/8/85	Nuclear submarine at Chazhma Cove, USSR	A spontaneous reaction during refuelling resulted in the destruction of the reloading house, damage to the submarine and the release of at least 10^{12} Bq of radioactive material. See section 3.1.3.	see section 3.3
26/4/86	Chernobyl NPP, USSR	A major accident released $3x10^{18}$ Bq of radioactive material into the atmosphere. See section 3.1.3 for more details.	see section 3.3
2/12/86	SILOE, Grenoble, France research reactor	The bottom of the pool was found to have degraded allowing water to leak out. There was a slight tritium contamination of underground water.	IAEA 1994
1986	Webbers Falls, Oklahoma, USA	An explosion of a tank containing radioactive gas at a uranium enrichment plant. One person was killed, eight injured.	Medvedev 1989
Sept 1987	Goiania, Brazil	Four people were killed, 6 seriously injured and 250 contaminated when a ^{137}Cs source was dismantled and sold as scrap. In order to help locate all the fragments an airborne survey using a helicopter at an altitude of 40m was conducted.	Berreto & Fonseca 1988 Pasquier 1996 Brown et. al.1996

2/12/91	EWA, Poland	A pressure container of Xe, used to produce ^{125}I, burst. About $5x10^6$ Bq of ^{125}I was released to the environment.	IAEA 1994
24/3/92	Sosnovyy Bor NPP, Russia	The accident resulted in the release of a small quantity of radioactive material, including small uranium fuel particles, that were detected in Finland.	Toivonen et. al.1992
6/4/93	Tomsk-7 reprocessing plant, Siberia	The accident released about $2x10^{13}$ Bq of radioactive materials. About 100 km^2 was severely contaminated, with fission products being detected as far away as Alaska.	Vakulovski et. al.1994 Larsen et. al.1994

Table 3.1: Accidents at nuclear facilities involving off-site releases. Further details of selected accidents are given in section 3.3, as indicated in the table. Details of other accidents, not involving off-site contamination, are given in Appendix B.

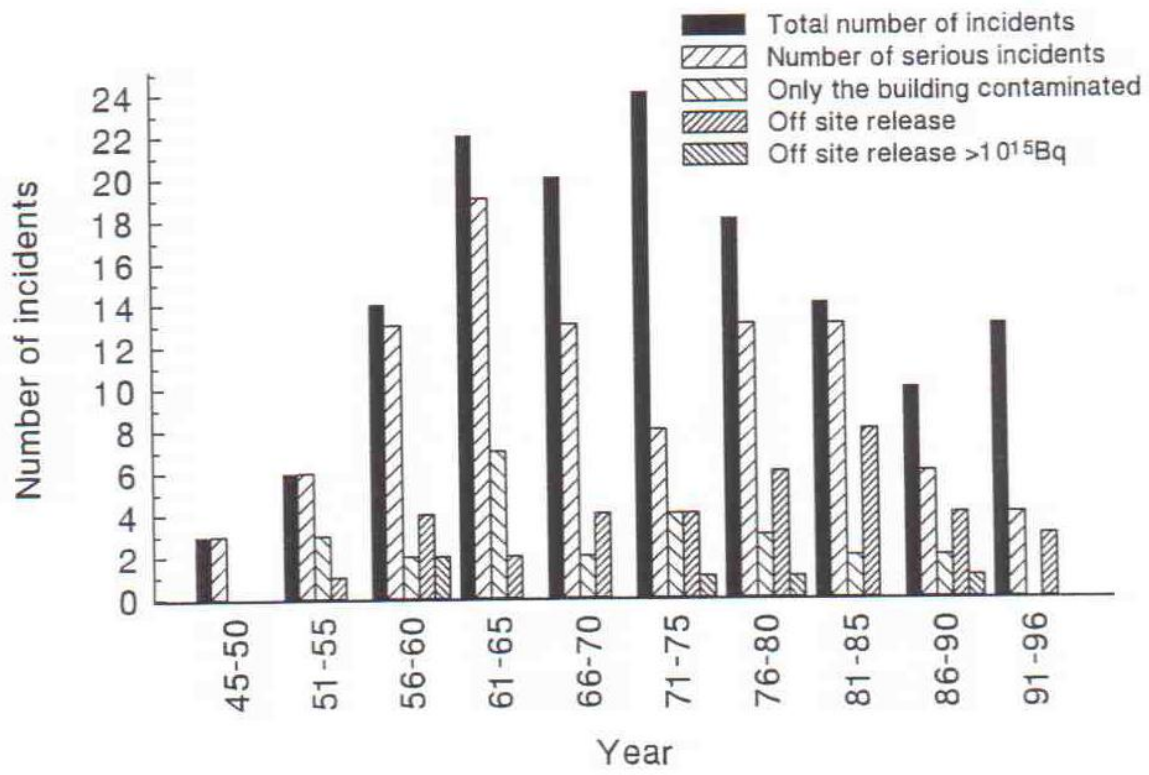

Figure 3.1: The distribution of nuclear incidents over five year periods. The plot shows the total number of incidents, the number of major incidents (including those in other categories in this plot), the number of accidents resulting in contamination of just the building, the number of accidents involving an off-site release and the number of accidents involving a major release.

3.3 Discussion of selected accidents

Accident at Chelyabinsk, Southern Urals, USSR, September 29th 1957.

The accident occurred at a military installation, located near the town of Kyshtym in the Chelyabinsk region of the former USSR, dedicated to producing plutonium for nuclear weapons, and is described in Nikipelov et. al. 1989, Romanov et. al. 1991 and Trabalka & Auerbach 1991. Waste from the plant was stored on a long-term basis in metal tanks encased in concrete, with the heat generated by radioactive decay dissipated by a water cooling system. In 1956, more than a year before the accident, corrosion and a failure of monitoring equipment led to a breakdown in the cooling system of a 300m^3 tank. The design of the tank, and the high radiation field, prevented repair of the system. This allowed the 70-80 tonnes of liquid wastes stored there, mainly as nitrate and acetate compounds, to heat up and dry out. At 4.20 pm, local time, on the 29th September 1957 the tank exploded with a force estimated as between 70 and 100 tonnes of TNT, the explosion being the result of a reaction between the acetates and nitrates in the drying waste.

The explosion ejected about $7x10^{16}$ Bq of radioactive fission products into the atmosphere to a height of approximately 1000m. The fall out from this cloud, which was blown in a north-easterly direction contaminated parts of three provinces, with the plume depositing over 0.1 Ci km^{-2} ($3.7x10^3$ Bq m^{-2}) of ^{90}Sr travelling some 300 km and covering an area in excess of 15 000 km^2. The area with deposition of ^{90}Sr greater then 2 Ci km^{-2} ($7.4x10^4$ Bq m^{-2}) covered some 1000 km^2 in a track 105 km long and 8-9 km wide, and a 120 km^2 area with a population of some 2000 received a deposition of ^{90}Sr in excess of 100 Ci km^{-2} ($3.7x10^6$ Bq m^{-2}). Figure 3.2 shows the distribution of this fallout, where it clearly follows a gaussian-like plume except for a small bulge at the south west end which is due to a 1967 release from dried radioactive sediments blown off the beach of a lake by a tornado.

Table 3.2 shows the composition of the radioactive material released. A noticeable feature of this composition is the small quantity of ^{137}Cs present, this was a major component of the Windscale and Chernobyl releases, and has a half life of 30.2 years. This is the result of the method used to concentrate these wastes, which was by means of precipitation with NaOH. The resulting precipitate contained practically all the radionuclides except the Cs isotopes which remained dissolved in the alkaline solution and were later concentrated separately. The ^{95}Zr-^{95}Nb content, amounting to some 20-25% of the total, is also somewhat surprising in waste that has been recycled and stored for over a year since the half-lives of these nuclei are relatively short (64 and 35 days respectively).

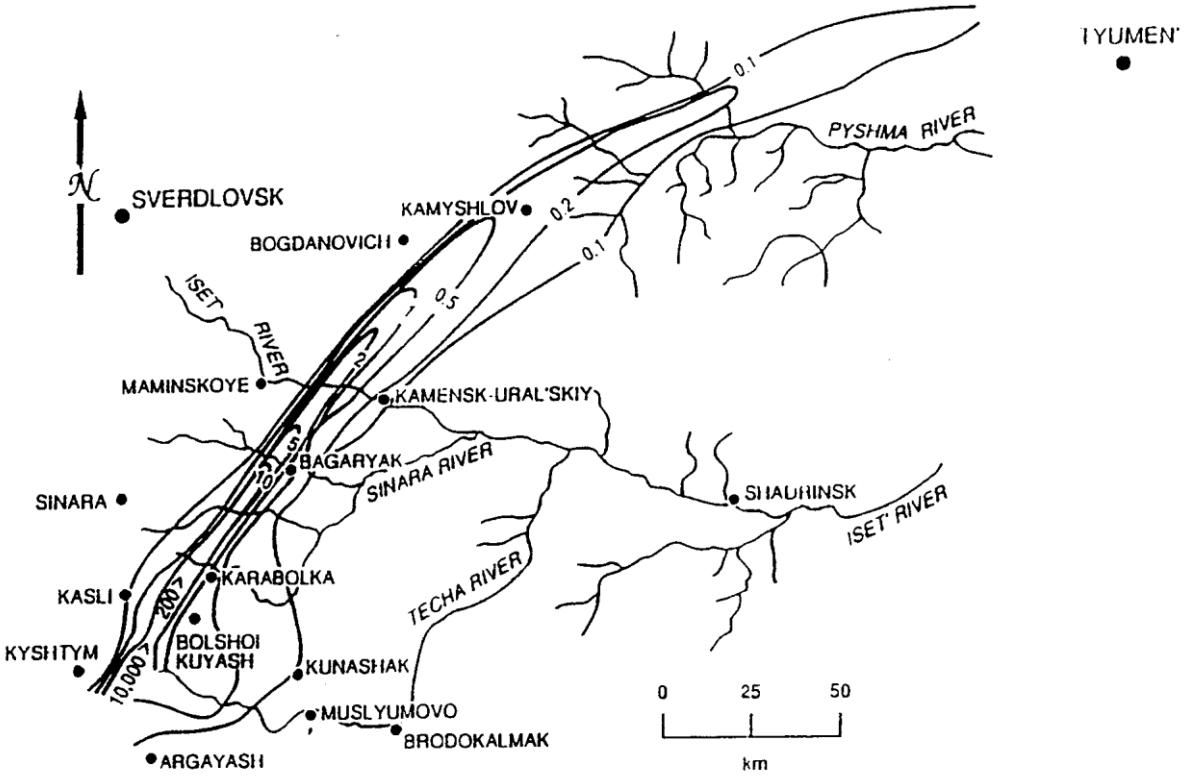

Figure 3.2: Deposition pattern of ^{90}Sr released from the Chelyabinsk plant. The contours are in units of Ci km^{-2} (1 Ci km^{-2} = 3.7 x 10^{4} Bq m^{-2}). Taken from Trabalka & Auerbach 1991.

Radionuclide	Activity released (x10^{15} Bq)	
	Nikipelov et. al.1989	Burnazyan 1990
^{89}Sr	traces	1.9
^{90}Sr+^{90}Y	4.0	5.2
^{95}Zr+^{95}Nb	20	16
^{106}Ru+^{106}Rh	2.8	2.6
^{137}Cs	0.03	0.07
^{144}Ce+^{144}Pr	48	47
remainder	traces	<9

Table 3.2: The composition of the material released from the accident at Chelyabinsk, USSR, September 29th 1957.

Accident at Windscale, UK, October 10th 1957.

The accident occurred in a graphite pile, used to produce plutonium for the British nuclear weapons programme, during a routine release of Wigner energy (energy stored in the graphite crystal lattice due to the displacement of carbon atoms by neutron bombardment), and is described in the official report (Atomic Energy Office 1957) and other sources including Arnold 1992. This was normally achieved by shutting down the pile, stopping the coolant flow and applying nuclear heating which raised the temperature of the pile sufficiently to anneal out the radiation damage, at which point the release was self-sustaining. The reactor was shut down at 7pm on the 7th October for such a procedure, but the graphite temperature was thought to be dropping and so a second application of nuclear heating was initiated from 11am on the 8th. With hindsight this second heating was unnecessary. Graphite temperatures continued to rise and air cooling was resumed at 10pm on the 9th of October. By the following morning stack activity had increased and visual inspection revealed that uranium was red hot in the vicinity of a thermocouple channel. By that evening, despite discharging fuel, some 150 channels were overheating. Attempts to cool the fire with carbon dioxide were unsuccessful, however it was subdued with water on the 11th and cooled the following day.

The fire released radioactive fission products totalling some 10^{15} Bq, excluding noble gases, through the 120m high stack. Wind initially carried the cloud of discharged materials towards the north-east beyond Penrith, later winds carried it south-easterly over Cumbria and Lancashire. Figure 3.3 shows the deposition of ^{131}I in NW England following the accident. The cloud was eventually carried into Scandinavia and Northern Europe. The peak discharge rates occurred at approximately midnight on the 10th and shortly afterwards, when the uranium was burning, and between 9 and 11am on the 11th, when water was first put on the fire and steam carried radioactive particles up the stack.

The composition of the radionuclides released, taken from several sources, is given in table 3.3. The values given by Clarke 1974 were derived from a computer inventory calculation with release factors of 0.1%, 1% or 10% for non-volatile, semi-volatile and volatile elements respectively. The inventory calculation required some assumptions about the reactor, such as the weight of the fuel, power rating and irradiation history, to be made since these were not given in the literature. The activities of daughter nuclides in equilibrium with their parents were assumed to be the same as the parents, and so were not given but have been included in table 3.3. The calculation included a three day cooling off period prior to the accident, during which the Wigner release took place. This resulted in the absence of very short lived radioisotopes in the release, with the exception of those in equilibrium with longer lived parents. The absence of ^{133}I is, however, puzzling since with a half life of 20.8 hours and a cumulative fission yield of 6.7% this should still be fairly common in the reactor inventory after three days. The activity due to its daughter, ^{133}Xe, is included, however. The earlier values of the radionuclide release were based on various measurements made shortly after the accident.

The various sources give some variety in the composition of the release, but they all agree within a factor of 10. The noble gases comprised some 67% of the release, a further 27% of the release was tritium. Of the remaining 6% of the release the largest components were ^{131}I

and ^{132}I, resulting from the decay of ^{132}Te. The tritium released was produced by irradiating rods of lithium-magnesium alloy, for research and the development of hydrogen bombs. ^{210}Po, used to initiate the explosive chain reaction in nuclear weapons, was produced in the reactor by the irradiation of bismuth oxide.

It is notable that the estimated ^{137}Cs release from Windscale, which depends mainly on the fuel age, varies in the published literature by a factor of 2, and that ^{137}Cs to ^{131}I ratios vary from about 3% to 7.7% depending on the source of the information used. This is of some interest, since the majority of environmental observations at the time of the accident referred to ^{131}I, although the ^{137}Cs output has been the subject of more recent interest following post-Chernobyl livestock movement restrictions in West Cumbria.

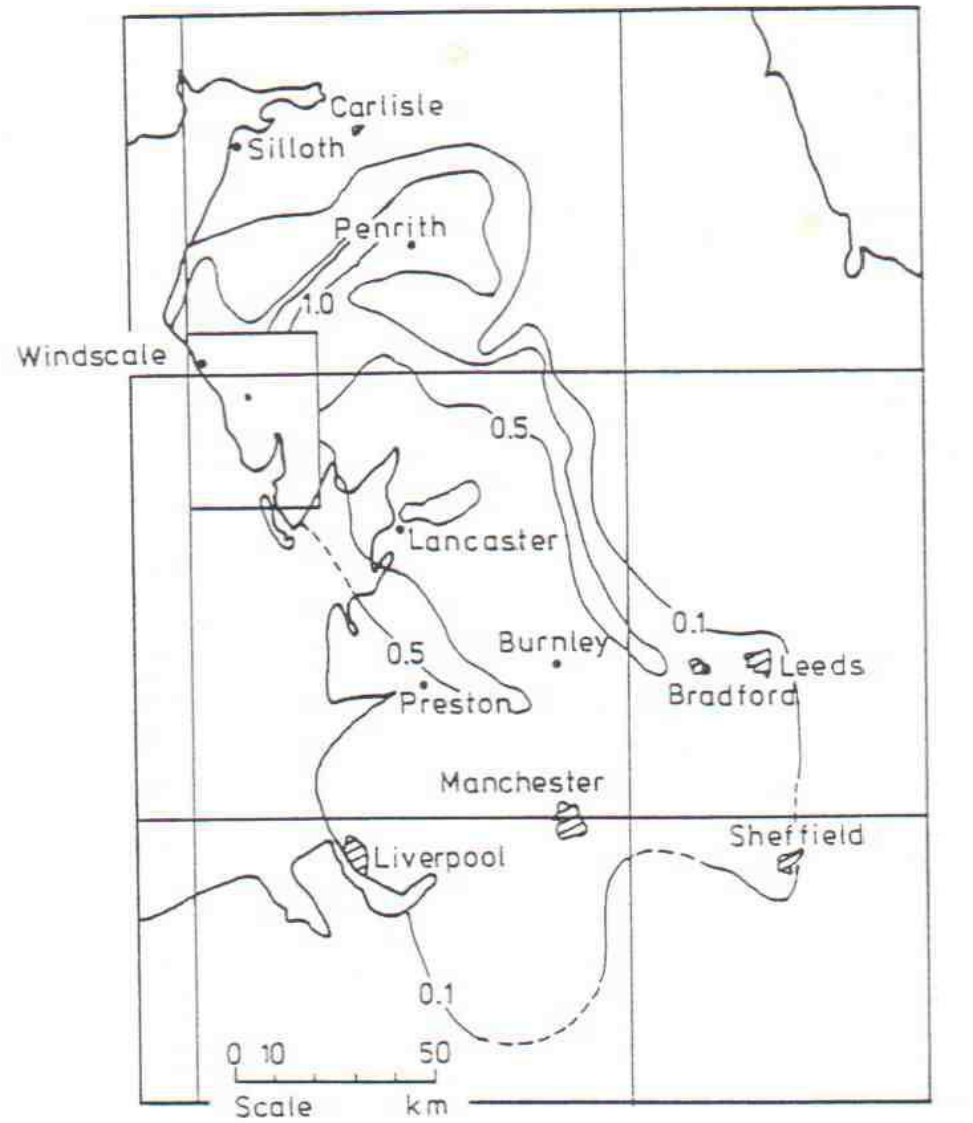

Figure 3.3: The deposition of ^{131}I in NW England following the Windscale fire, the activities are in $\mu Ci\ m^{-2}$ ($1\ \mu Ci\ m^{-2} = 3.7 \times 10^4\ Bq\ m^{-2}$). Taken from Chamberlain 1959.

Radionuclide	Estimated radionuclide release ($x10^{12}$Bq)					
	Dunster et.al.(1958)	Loutit et.al. (1960)	Beattie (1963)	Clarke (1974)	Chamberlain (1981)	Crick and Linsley (1984)
^{3}H						5 000
^{85}Kr				59		59
^{89}Sr	3.0	3.0	3.0	5.1	7.4	2.5
^{90}Sr	0.33	0.074	0.074	0.22		0.06
^{90}Y *				0.22		0.06
^{91}Sr				0.037		0.037
^{91}Y				6.4		6.6
^{93}Y				0.059		0.059
^{95}Zr				7.5		7.8
^{95}Nb *				7.5		7.8
^{97}Zr				0.36		0.36
^{99}Mo				36		36
^{103}Ru				40		40
^{105}Rh				0.52		0.52
^{106}Ru		3.0	3.0	5.9		2.5
^{106}Rh *				5.9		2.5
^{111}Ag				0.52		0.52
^{115}Cd				1.6		1.6
^{123}Sn				0.24		0.24
^{125}Sn				0.25		0.25
^{127}Sb				0.033		0.034
^{129m}Te				25		25
^{129}Te *				25		25
^{131m}Xe				65		66
^{131}I	740	740	740	600	1 000	600
^{132}Te		440	440	600		354

^{132}I *				600		
^{133}Xe				12 000		12 600
^{134}Cs				1.2		1.2
^{135}Xe				35		35
^{136}Cs				1.5		1.5
^{137}Cs	22	22	22	46		18
^{140}Ba				6.4		6.6
^{140}La *				6.4		6.6
^{141}Ce				7.1		7.2
^{143}Ce				1.6		1.6
^{144}Ce		3.0	3.0	4.0		2.5
^{147}Nd				2.3		2.3
^{147}Pm *				2.3		2.3
^{149}Pm				0.59		0.59
^{151}Pm				0.10		0.10
^{153}Sm				0.078		0.078
^{156}Eu				0.26		0.26
^{210}Po						8.8
^{235}U						2.0×10^{-6}
^{238}U						6.0×10^{-5}
^{239}Pu						1.6×10^{-3}

Table 3.3: The composition of the activity released from the accident at Windscale, UK, October 10th 1957. The nuclei labelled * are short lived daughters in equilibrium with longer lived parents.

Crash of Cosmos-954, January 24th 1978.

Cosmos-954, a nuclear powered Soviet satellite, re-entered the Earth's atmosphere and broke up over Canada's Northwest Territories. The resultant satellite fragments were spread along an east-west trajectory, covering an area approximately 800km long and 50km wide, although some minute particles were collected up to 400km south of this track as a result of dispersion by the wind. The radionuclides found included ^{54}Mn, ^{58}Co (activation products), ^{95}Zr, ^{95}Nb, ^{103}Ru and ^{140}La. The more volatile elements (eg: I and Cs) and noble gases burnt off during re-entry and so were no longer present in the fragments which reached earth. The accident is described in Bristow 1978 and Grasty 1980.

Accident during submarine refuelling at Chazhma Cove, USSR, August 10th 1985.

The accident occurred in a nuclear-powered submarine moored at a dockyard in Chazhma Cove for repairs, and is described in Sivintsev et. al. 1994. After the reactors were refuelled a spontaneous chain reaction occurred in the starboard reactor as a result of a violation of nuclear safety rules. In the resultant thermal explosion the bow and stern equipment compartments and the reloading house were destroyed (the roof of the reloading house was thrown 70-80m), the pressure hull in the stern part of the submarines' reactor bay was damaged, and fresh fuel was thrown out of the reactor. A fire in the reactor bay took four hours to contain. Particles of fuel, fission fragments and activation products, totalling approximately 10^{13} Bq, were precipitated around the submarine with a radius of 50-100m, with a surface density of (80-700)x10^{6} Bq m^{-2}.

Accident at Chernobyl, USSR, April 26th 1986.

The accident occurred during an experimental test of the reactors' emergency systems, and is described in several sources including INSAG 1986, Gittus et. al. 1987 and Medvedev 1989. The experiment started at 1am on April 25th by reducing power in the reactor by 50%, from 3000 to 1500 MW(th), where it was held from 1pm. At 11pm power reduction was resumed towards a target of 700-1000 MW(th), but operator error led to the power reduction continuing to 30 MW(th). The reactor was stabilized at 200 MW(th) (below the minimum permissible power level of 700 MW(th)) at 1am on the 26th. At this power the reactor had a positive void coefficient, and several safety systems had been shut down and too many control rods removed. These mistakes left the reactor in a very unstable state, and when the experiment was initiated by tripping the turbine the only significant heat sink in the reactor was removed. The result was boiling of the coolant, resulting in an uncontrollable rise in reactivity reaching an estimated 100 times full power. This power excursion resulted in a substantial and sudden increase in pressure within the reactor, rupturing a number of fuel channels. The reactor space had been designed to withstand the rupture of a single fuel channel, and it now over-pressurized and the upper plate weighing about 1000 tonnes was lifted. This ruptured all the remaining fuel channels, lifted the control rods and sheared pipes. A second explosion occurred 2 to 3 seconds later which destroyed the reactor building. Upon exposure to air the graphite moderator caught fire, and chunks of burning graphite and fuel material were thrown from the reactor building, starting secondary fires that threatened the other reactors on the site.

After heroic efforts by fire fighters these secondary fires were extinguished before any damage to the other reactors or their control systems could occur. The damage to the building allowed an air flow through the core to develop, fanning the fire there. The operators succeeded in injecting water into the reactor building, but most of it flowed out of the reactor towards units 1 and 2, where it threatened to short circuit the control systems, and water flow was stopped after about half a day. The fire burnt for nine days, and was eventually extinguished by dumping about 5000 tonnes of various materials (mostly sand and clay, along with boron carbide and lead) into the reactor well from helicopters, and pumping nitrogen through the reactor to reduce the core temperature and starve the fire of oxygen. Approximately 135 tonnes of fuel (about 71% of the total fuel in the reactor) melted and flowed into the lower regions of the reactor where it eventually solidified into various kinds of lava-like materials. After initial clean-up operations, the remains of the reactor building were enclosed in a concrete and steel sarcophagus, the long term condition of which remains unclear.

The accident resulted in the release of approximately 100% of the noble gases, 10-20% of the volatile radionuclides and 3-6% of the more refractory radionuclides present in the core, a total release of about $3x10^{18}$ Bq. About 25% of this release occurred on the first day of the accident, the rest was released over a period of nine days. Some 3-4% of the fuel in the core was also expelled from the reactor; 0.3-0.5% was deposited within the site, 1.5-2.0% was deposited within 20km of the site, and 1.0-1.5% was deposited beyond 20km of the site. About 50% of the released material was deposited within 30km of the reactor. The composition of the active material deposited close to the reactor was similar to that of whole fuel, enhanced in volatile fission products by a factor of 3-6. Samples collected in western Europe were composed almost entirely of volatile fission products, with only a few percent composed of less volatile products and fuel (possibly released as fine aerosols in the initial explosion).

Initially the activity released was transported by the wind in a north-westerly direction over Scandinavia. A few days later the wind shifted, transporting the activity in a more westerly direction. A high pressure system that moved easterly across Europe resulted in a wide dispersal of the material. Figure 3.4 shows the distribution of ^{137}Cs near Chernobyl, figure 3.5 shows γ-dose rates in the European part of the former USSR. The pattern of deposition was very patchy, largely reflecting rainfall as the cloud passed.

The composition of the radionuclide release is given in table 3.4. About 60% of this release was due to noble gases, indicating a much larger proportional release of other radionuclides than in the Windscale fire. Of the remaining 40% of the release the largest component was ^{131}I, as in the Windscale fire, but the proportion of less volatile radionuclides was much larger. This difference reflects the much greater temperatures in the Chernobyl accident, in which the majority of the fuel completely melted, than at Windscale where the only some of the fuel elements slightly melted. The greater temperature resulted in a greater release of the more refractory elements from the reactor.

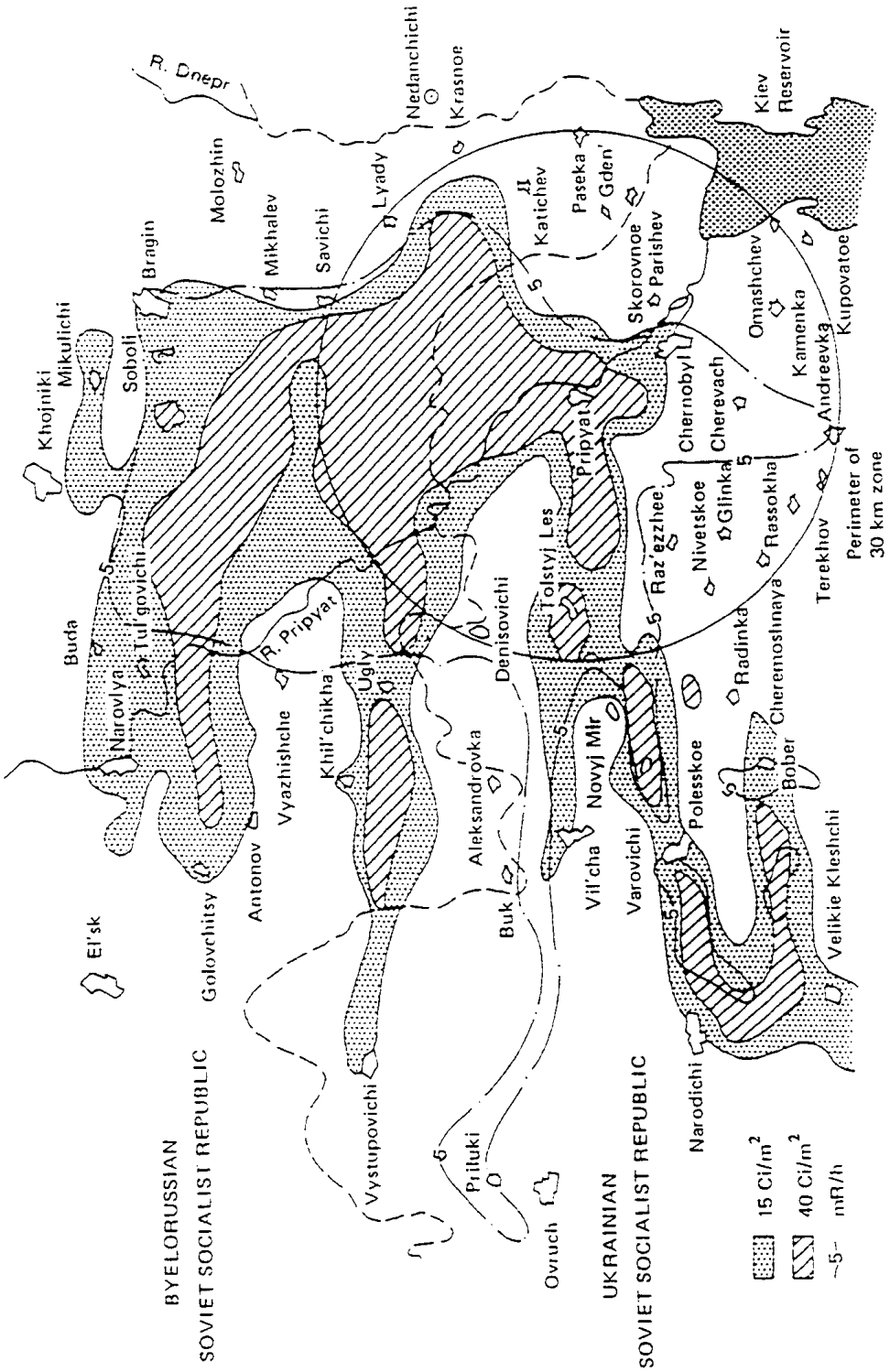

Figure 3.4: The distribution of ^{137}Cs deposition near the Chernobyl site (10th May 1986), taken from Izraehl' 1990. 1 Ci m^{-2} = 3.7x10^{10} Bq m^{-2}.

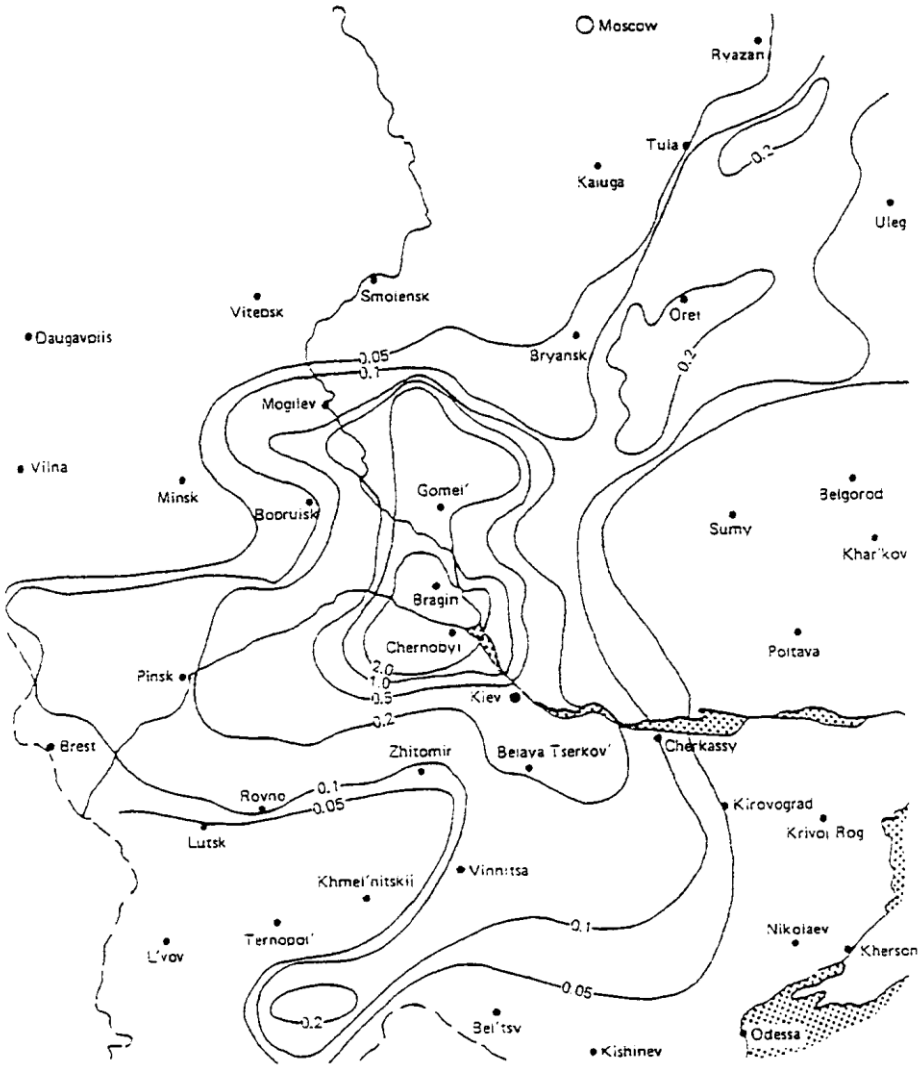

Figure 3.5: Map of the γ-dose rate (mR/h) in the European part of the former USSR (late May to early June 1986), taken from Izraehl' 1990.

Radionuclide	Released activity ($x10^{15}$ Bq)				
	INSAG 1986	Gudiksen et. al. 1991	Buzulukov & Dobrynin 1993	Sich et. al. 1994	Devell et. al. 1995
^{85}Kr	33	≥18	33	33	
^{89}Sr	74	22	81	81	~115
^{90}Sr	7.4	1.3	8.1	8.0	~10
^{91}Y		7.2			
^{95}Zr	140	8.5	167	170	196
^{99}Mo	110	17		210	>168
^{103}Ru	120	27	170	170	>168
^{106}Ru	60	6.3	30	30	>73
^{127}Sb		18			
^{127m}Te		3.7			
^{129m}Te		13		240	
^{131}I	260	1 300	1 700	1 700	~1760
^{131m}Te		6.7			
^{132}Te	48	200	400	1 000	~1150
^{133}I		300			
^{133}Xe	1 700	≥4200	6 300	6 500	6 500
^{133m}Xe		≥86			
^{134}Cs	20	48	44	44	~54
^{136}Cs		20			
^{137}Cs	37	89	85	85	~85
^{140}Ba	160	37	170	170	~240
^{140}La		10			
^{141}Ce	100	8.5	200	200	196
^{144}Ce	90	5.2	140	140	~116
^{147}Pm		0.9			

^{238}Pu	0.03		0.03	0.03	0.035
^{239}Pu	0.03		0.03	0.03	0.03
^{239}Np	4		170	1 700	945
^{240}Pu	0.04		0.04	0.04	0.042
^{241}Pu	5		6.0	5.9	~6
^{242}Cm	0.7		0.9	0.9	~0.9

Table 3.4: The composition of the activity released from the accident at Chernobyl, USSR, April 26th 1986. The values given in the INSAG 1986 report are decay corrected to May 6th and have uncertainties of 50%. The values given by Gudiksen et. al.1991 are decay-corrected for three days after the accident and have uncertainties of 30%. The remaining estimates are decay corrected for April 26th.

3.4 General Features of Source Terms

For accidents involving nuclear reactors or reprocessing plants the source term may be complex, but has some general features that result from the fuel history, the temperature during the accident and the state of containment. The source terms for other accidents are generally much simpler, for example, a lost source may consist of just one radioisotope, which may already be known. Similarly potential source terms for nuclear weapons accidents will generally be known.

The following sections examine some of the factors which influence reactor accident source terms.

3.4.1 Dependence of source terms on fuel history

For an accident at a reactor, the history of the fuel, in particular its age, and the power of the reactor in the hours or days prior to the accident, determines the radionuclide inventory and thus constrains the release term. Unirradiated fuel comprises fissile and fissionable actinides, mainly α-emitters of low specific activity, and their immediate decay products. After short periods of irradiation in the reactor these are supplemented by fission products produced in proportion to their fission yields. With prolonged irradiation the quantities of short lived fission products progressively saturate, as production and decay rates equalise. Longer lived products and stable fission products grow in from isobaric decay chains, and transuranic nuclides and other activation products are produced. In this manner the composition of the radioactive inventory of the fuel changes, while its specific activity generally increases. Irradiation history therefore determines the activities which might be affected by accidents and partly released.

Post-irradiation delays result in preferential loss of shorter lived nuclides, thus providing one mechanism for simplification of the source term. Power variations during irradiation also

influence the relative proportions of short and long lived nuclides, and of products with high neutron capture cross sections whose concentrations depend on burn-up.

In the case of the Windscale fire the accident involved relatively young fuel and the power had been reduced 2-3 days prior to the start of the Wigner release, thus simplifying the source term in relation to short lived fission products. For the Chernobyl accident the power was reduced to 50% of normal operating levels 12 hours before the accident, and to about 10% 4 hours before the accident. This shorter low power period and the larger, older fuel load resulted in the larger, more complex source term.

The material released at the Kyshtym accident had been stored for over a year, allowing the decay of shorter lived fission products.

3.4.2 Dependence of source terms on temperature

The temperature of the core during an accident is an important parameter in determining the source term. The thermal history of the accident, together with the chemical and physical composition of material in the core, determines the proportions of the active inventory which are mobilised for potential release. The outcome will depend upon the volatility of the elements and compounds present, release from the core proceeding in order of noble gases, volatile components (eg: iodine nuclides) and less volatile materials in order of increasing temperature. The nature and state of fuel cladding, and the physical form of the fuel are also significant.

Core temperature played a significant part, for example, in differentiating the release of noble gases and volatile elements at Three Mile Island from the wider range of nuclides, including refractory fission products, and actinides at Chernobyl. In the case of the Cosmos-954 satellite crash volatile elements burned up during re-entry, the solid material reaching ground level thus comprising predominantly less volatile nuclides.

The temperature of the core also affects the dispersion of the radionuclides once they are released, particularly if there is a complete loss of containment. A very hot core will result in the particles and gases released being carried further up into the atmosphere as a result of buoyancy effects, from where they will be carried further before being deposited on the ground.

3.4.3 Dependence of source terms on containment

Another factor determining the source term for a nuclear accident is the state of the reactor containment during and after the accident. For reactors the containment comprises the fuel cladding, the reactor vessel and in some cases a containment building, each of which acts as a barrier to accidental release. If the containment is intact then the release of radionuclides into the environment will be small. On the other hand, an accident in which the containment is destroyed will result in the release to the environment of all the radionuclides that are released from the core. If the containment fails some time after the accident there will be a smaller

release of radioactive material as a result of the decay of short lived nuclides and the plate-out of particulates and adsorbed species inside the containment building.

In de Boeck 1993 three classes of containment failure are considered; early failure, late failure and containment bypass. An early containment failure is a failure of the containment prior to or shortly after core debris enter the reactor vessel. Possible early failure mechanisms include direct containment heating in which the heat from core debris causes the containment vessel to overpressurise, steam explosions in which steam generation results in a shock wave, hydrogen combustion or isolation failures. Late containment failures are failures of the containment in the longer term. Possible late failure mechanisms include late burning of combustible gases, gradual overpressurisation, basemat meltthrough and overheating. Containment bypass failures are accident scenarios in which the containment is completely bypassed, for example when primary coolant escapes into secondary buildings without first being discharged into the containment volume.

The state of the containment will also affect the distance over which radionuclides are deposited. This is a result of a containment building increasing the time it takes for the radionuclides released from the core to reach the atmosphere, by which time the air transporting them will have cooled. Cooler air will not rise as far into the atmosphere when it gets outside the containment building, and so the radionuclides will be deposited closer to the accident site.

In the case of the Windscale fire filtration was thought to have intercepted approximately 60% of the nuclides (Arnold 1992). At Three Mile Island the containment building made a critical difference to the radiological outcome. The absence of effective containment to deal with the forces of the Chernobyl blast had far reaching consequences for the release.

4. GAMMA-RAY SPECTRA

4.1 AGS Systems

Airborne gamma-ray survey (AGS) systems consist of a γ-ray detector, most commonly a high volume sodium iodide (NaI) system, mounted on an aircraft. The response of the system depends on the ability of γ-rays to penetrate a few hundred metres in air. Thus, an aircraft flying at an altitude of 100m or less can readily measure spectroscopic information from γ-emitting radionuclides on, or just below, the surface. Analysis of such spectra allows the total γ-dose rate and the activities of some individual radionuclides to be determined. By linking the spectrometer with positional information (eg: from a GPS system) maps of the distribution of activity can be produced. The use of computers allows the spectra to be analysed rapidly, enabling such maps to be produced shortly after the aircraft has landed.

AGS systems were initially developed for geological purposes, in particular uranium exploration. However with improvements to detectors, electronics and computing facilities they have also proven useful in environmental surveys. AGS has been used to map natural radioactive backgrounds for the purpose of epidemiological studies of cancer, studies of discharges from nuclear facilities, mapping of fall-out from past nuclear accidents and weapons tests, and as part of preparations for nuclear emergency responses.

The advantages of AGS systems are the speed of data collection and presentation, and the large areas that can be easily and rapidly mapped. The data collected, being the total activity present in a relatively large area, avoid problems with sub-sampling errors (where a small sample area may not be representative of the whole local environment). The limitations of current AGS systems include the relative lack of sensitivity to radioisotopes that emit only low energy γ-rays and the poor energy resolution inherent in NaI detectors, resulting in spectral interferences in complex cases. It is possible to use Ge semiconductor detectors which have a much higher resolution, but their efficiency is considerably lower, limiting the statistical precision available in short integration times, except in highly active areas. Further details of the principles of AGS systems can be found in ICRU 1994 and IAEA 1991. Details of the SURRC AGS system can be found in Sanderson et. al. 1990a, 1995b.

4.2 Spectra Recorded From Past Accidents

Gamma-ray spectra were recorded following several accidents using aerial survey systems, in situ spectrometry and from a variety of samples. These spectra were recorded over extended periods of time after the accident, but for the present report, considering primarily short lived fission products, only those spectra recorded within one year of the accident will be considered.

Spectra recorded after the Windscale fire

An aerial survey around the Windscale site was conducted between the 19th and 22nd October 1957, Williams et. al. 1958. This survey consisted solely of a measurement of the total dose

rate using small volume NaI(Tl) detectors, recorded in-flight on a paper chart. Due to the equipment available, no spectrometric information was recorded at the time, so far as can be determined, either from the air or the ground.

Spectra recorded after the Crash of Cosmos-954

Because of the size of the area to be covered, the primary means of locating the fragments of Cosmos-954 was an aerial survey using γ-ray spectrometers mounted in four Hercules aircraft. The Geological Survey of Canada (GSC) spectrometer consisted of a 50 350 cm^3 NaI detector from which 256 channel γ-ray spectra were recorded. Additional flights, at a lower altitude, were conducted using detection equipment mounted on helicopters. These flights were used to more precisely locate fragments observed from the Hercules aircraft, and they were also more sensitive to small sources. When a source had been located by airborne methods, it was precisely located and removed by personnel on the ground. Approximately 3500 minute particles were eventually collected over a four month period.

Correction for the large natural background at the crash site was a major consideration, resulting in the need to use a ratio technique to distinguish between natural and artificial radiation sources, and is described by Bristow 1978. This involved taking the ratio of the number of counts in a window from 300-900 keV (due mainly to fission products) to the number of counts in a window from 900-1500 keV (due mainly to natural sources). Figure 4.1 shows the total count rate and this ratio along a flight path overflying the first fragment to be positively identified, the strong signal at 1.5 km is artificial but the weaker signal from 3 km is due to natural background. The ratio only picks out the artificial signal. This ratio technique made it possible to determine anomalous readings corresponding to artificial point sources on the ground, and hence locate fragments of the satellite.

A technique to remove the natural contribution to the spectra showing enhanced man-made activity was also devised, and is again described by Bristow 1978. This involved summing 20 natural spectra (10 from each side of the anomaly) and each of the spectra recorded over the anomaly is scaled by a factor of 20. The background is then subtracted from the anomalous spectra, and the result smoothed. Figure 4.2 shows the natural background and fission product spectra recorded with this equipment for the first fragment to be positively identified. The fission product spectrum shows the typical predominance of low energy γ-lines due to several fission products.

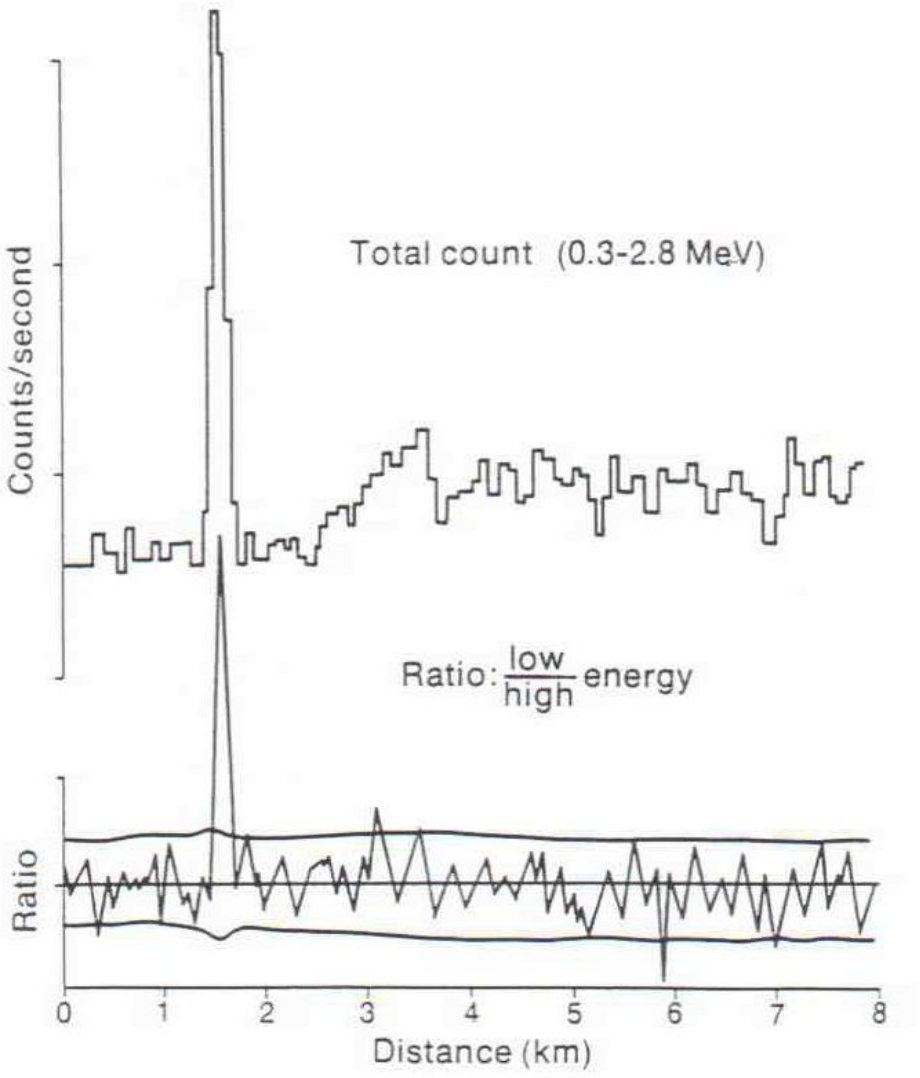

Figure 4.1: The total count profile for a flight line over the first confirmed fragment from Cosmos-954 showing the use of the ratio technique. Taken from Grasty 1980.

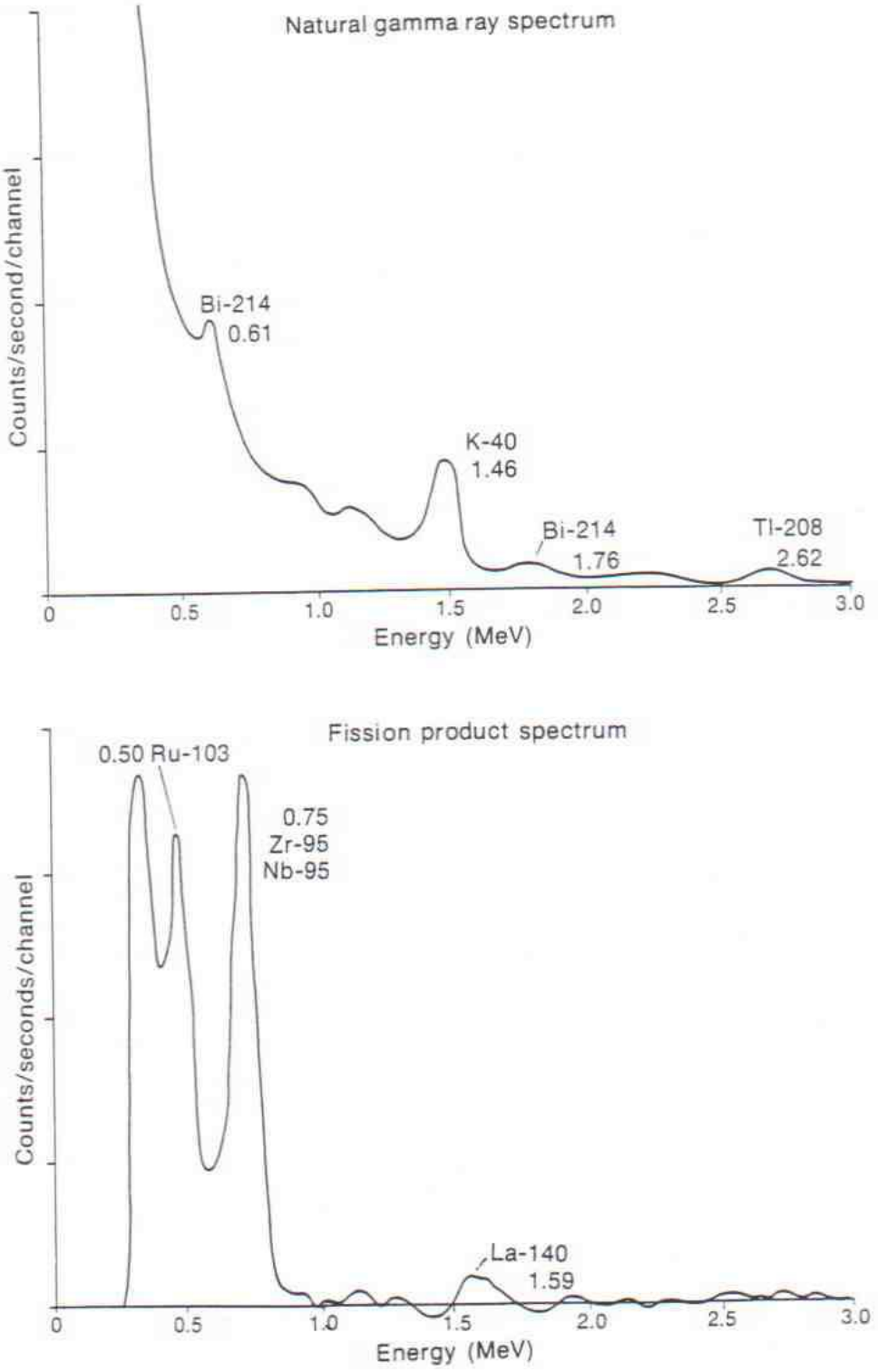

Figure 4.2: A typical natural γ-ray spectrum and fission product spectrum for the first confirmed fragment of the Cosmos-954 satellite, recorded with the GSC NaI based spectrometer. Taken from Grasty 1980.

Spectra recorded after the Chernobyl accident

Following the Chernobyl accident a very large number of radiological surveys were conducted, many of which used γ-ray spectrometry methods. These included measurements on samples of soil, plants, water, air filters and other materials. There were also a range of in-situ measurements, and surveys using detectors mounted on aircraft or ground vehicles. The following is a collection of some of the spectra recorded by these various methods up to about a year after the accident.

The first airborne γ-ray surveys following the accident were conducted in Sweden by the Swedish Geological Company (SGAB) in cooperation with the Swedish National Institute of Radiation Protection (SSI), and are described in Mellander 1989. The SGAB γ-ray spectrometer consisted of a 16.8 litre NaI(Tl) detector, although due to the high count rates only half of this was used initially, producing 256 channel spectra. This was mounted in the luggage compartment of an Aero Commander 680 twin engine fixed wing aircraft. The first survey of Chernobyl fall-out began on the afternoon of the 1st May 1986, 4 days after the radioactive cloud was first detected over Sweden. The whole of Sweden, except for the far north, had been mapped by the 8th May with east-west lines 100 km apart at an altitude of 150m. On the morning of the 9th May the aircraft returned south to map the fall-out from a new radioactive cloud (flying through the cloud on the way, contaminating the aircraft). Between the 9th and 23rd May the whole country was covered with 50 km line spacing. Figure 4.3 shows a spectrum measured in May 1986 with the 8.4l detector, and figure 4.4 shows a spectrum measured in June 1987 using the full 16.8l detector. These spectra show several peaks, corresponding to a large number of different γ-lines. The earlier spectrum shows peaks at approximately 550keV, 630keV, 780keV, 1500keV and 1600keV. The peak at 1500keV includes ^{40}K lines (natural background), the peak at 1600keV is due largely to ^{140}La. Initial caesium deposition maps were based on the 780keV signal, calibrated by ground to air comparisons, and assumed to be largely composed of ^{134}Cs. In the later spectrum many of these peaks have disappeared (for example the 1600keV La peak), and other peaks have become apparent including a 660keV peak due to ^{137}Cs.

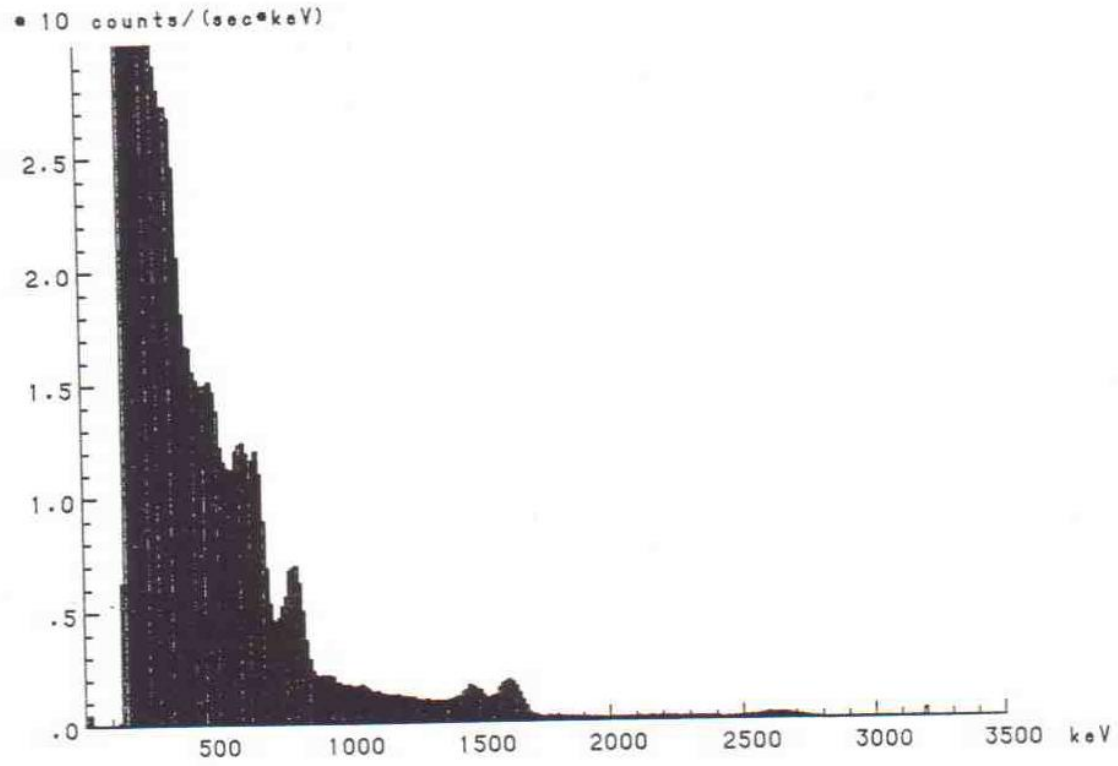

Figure 4.3: A γ-ray spectrum measured with the SGAB 8.4l NaI detector at an altitude of 150m in May 1986. Taken from Mellander 1989.

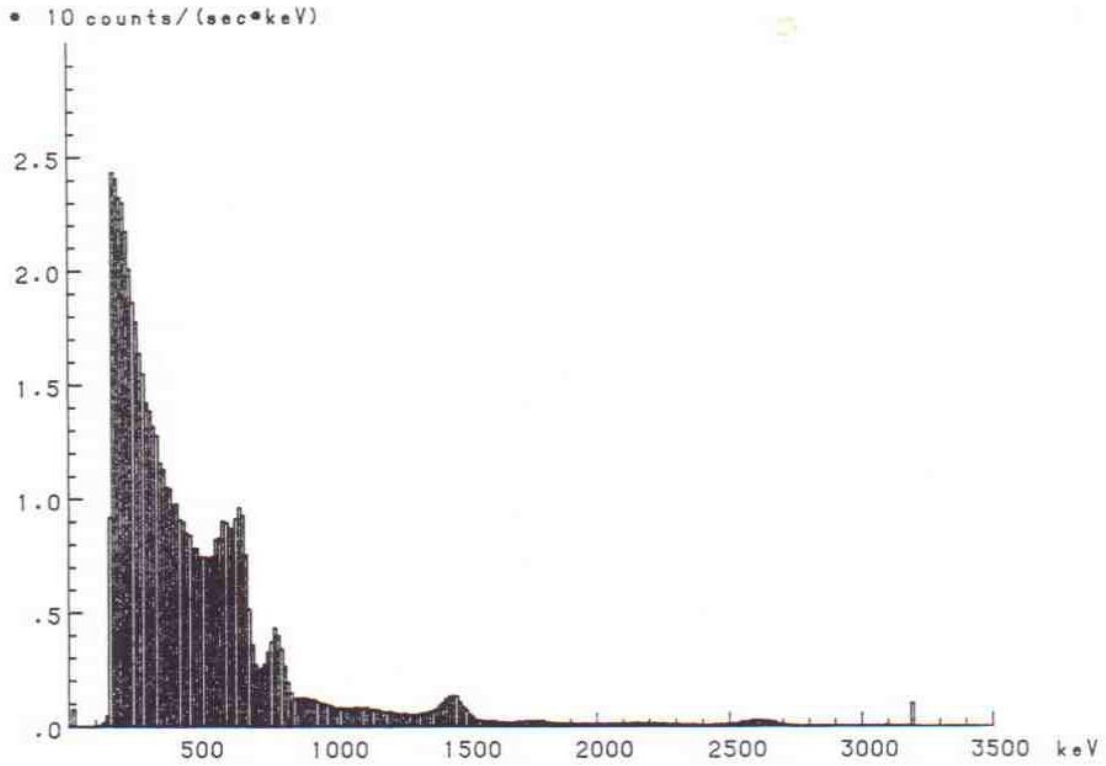

Figure 4.4: A γ-ray spectrum measured with the SGAB 16.8l NaI detector at an altitude of 150m in June 1987. Taken from Mellander 1989.

On the 29th April 1986 the survey aircraft of the Geological Survey of Finland (GSF) was returning from a survey in the north of Finland to Helsinki. Having heard on the radio that significant levels of radioactivity had been detected in Finland and Sweden they had turned on their spectrometer consisting of 25 litres of NaI, recording 120 channel spectra covering an energy range of 200 to 3100 keV using a 1 s livetime. During the course of this flight they flew through the radioactive plume from the Chernobyl accident, heavily contaminating the aircraft. Peaks attributed to iodine, caesium and lanthanum isotopes were visible (Multala 1995), although it has subsequently been realised that ^{132}I interferes with both Cs peaks. Figure 4.5 shows a spectrum recorded over a 30 minute period in the cloud (Grasty et. al. 1996.), importantly the ^{131}I peak is well resolved. This peak was not clearly resolved in the spectra recorded by the SGAB group in early May (figure 4.3), where it is within a much larger background due to the scattering of higher energy γ-rays in the atmosphere.

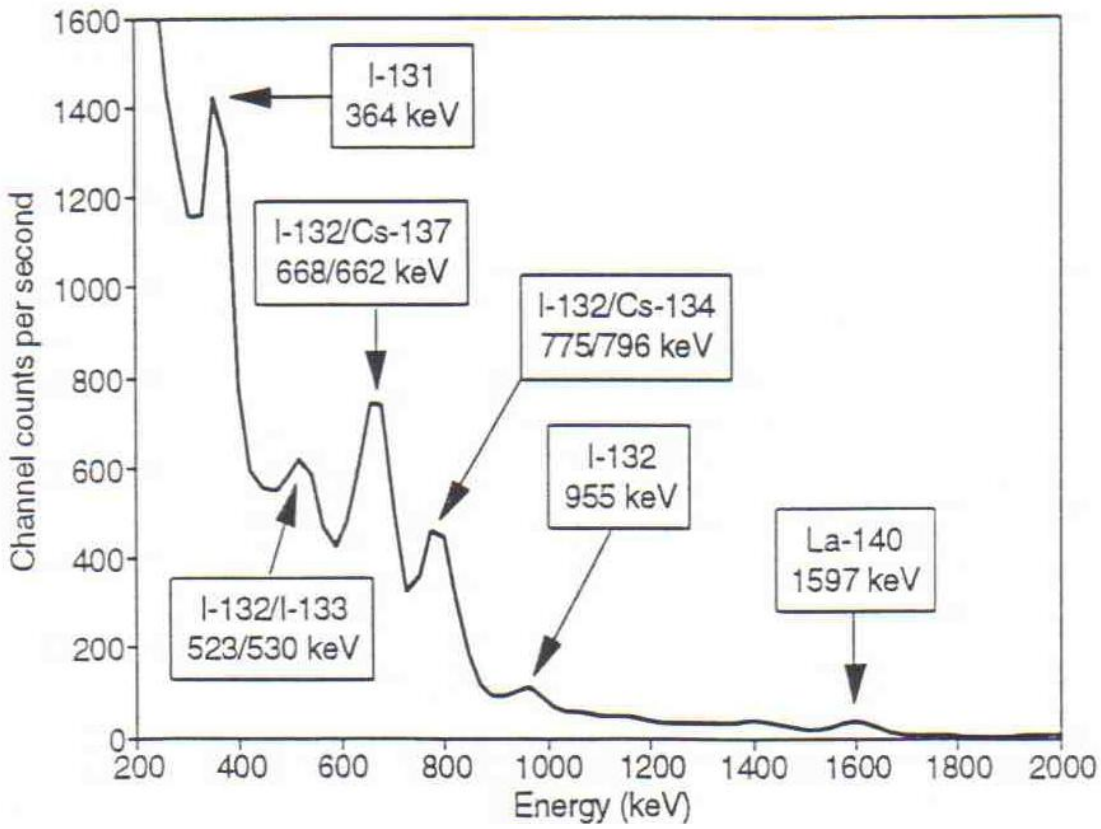

Figure 4.5: A γ-ray spectrum measured with the GSF NaI based spectrometer within the Chernobyl plume averaged over 30 minutes, taken from Grasty et. al. 1996.

An airborne survey of the highly contaminated areas of central Norway was conducted between 5th May and 6th June 1986 to complement a survey of the entire country conducted with car based spectrometers (Lindahl & Haabrekke 1986).

Airborne surveys around the site of the accident and in neighbouring regions were conducted from May 1986 (Stukin 1991 and Nagaoka et. al. 1994). These surveys utilised the "Makfar-2" spectrometric system and was conducted by the "Aerogeology" division of the Soviet Ministry of Geology. This system is composed of NaI(Tl) crystals arranged to produce total volumes between 1.25l and 48l, with on-board data storage and ground-based computer processing. The surveys covered a 5 km zone around the plant with a 100m line spacing, a 60 km zone with 500m spacing and five other regions with a 3km line spacing.

In addition to the airborne surveys, various measurements were also made using in-situ spectrometers. An example of such measurements were those made at the German Institute for Radiation Hygiene near Munich (Winkelmann et. al. 1987). Figure 4.6 shows spectra recorded, using a Ge detector with an integration time of 30 mins, before and after the Chernobyl accident. The peaks in the spectrum recorded before the accident shows a normal natural background from the Th series (^{208}Tl, ^{212}Pb and ^{228}Ac), the U series (^{214}Bi and ^{214}Pb), ^{40}K and the 511keV line due to electron-positron annihilation (the positrons are formed by pair production, in which a high energy γ-ray interacts with matter producing an electron-positron pair). The spectrum recorded after the accident shows a large number of additional γ-ray peaks superimposed on this background, some of which are identified or resolved from other peaks.

Samples of soil and rainwater were collected in many locations across Europe, several of which were analysed using γ-spectroscopic methods. Figures 4.7-4.11 show a selection of spectra from samples collected in Denmark (Hovgaard & Korsbech 1992) and at the Scottish Universities Research and Reactor Centre. Again, these spectra contain a large number of peaks from several radionuclei; in the NaI spectra these are composites of several different discrete γ-lines, and even with the Ge spectra some of them are not fully resolved. With the exception of the clay samples, these spectra only contain γ-rays from fission products due to the advantage of being able to shield the sample and detector from the environmental background. The spectra shown in figure 4.10 are from a clean clay sample, and the same clay sample contaminated with a small amount of the gutter mud used for the measurement in figure 4.9. This increased the total counts at energies below 900 keV by 6-7%.

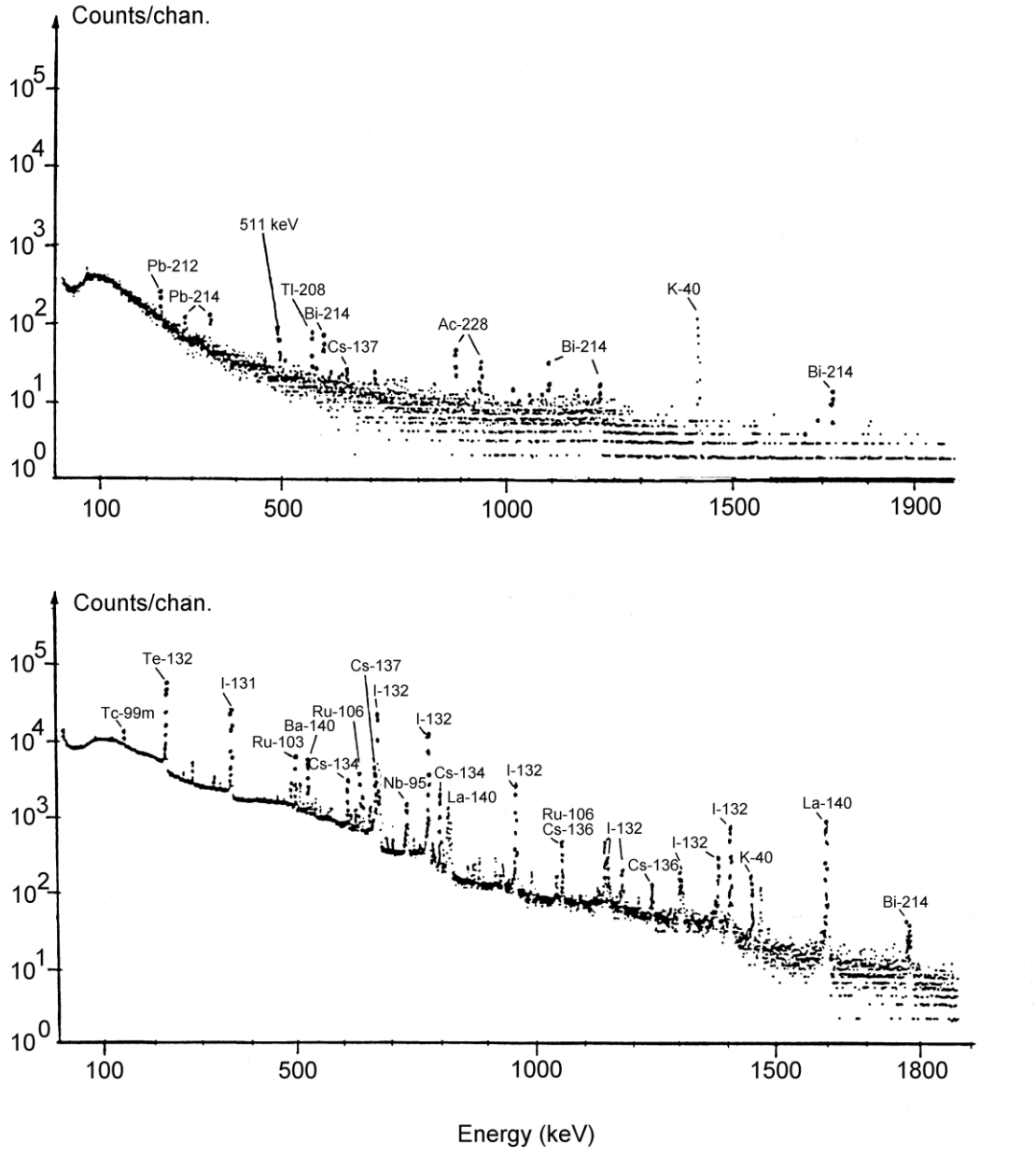

Figure 4.6: γ-ray spectra recorded with an in-situ Ge based spectrometer at the German Insitute for Radiation Hygiene before (top) and after the Chernobyl accident. Taken from Winkelmann et al 1987.

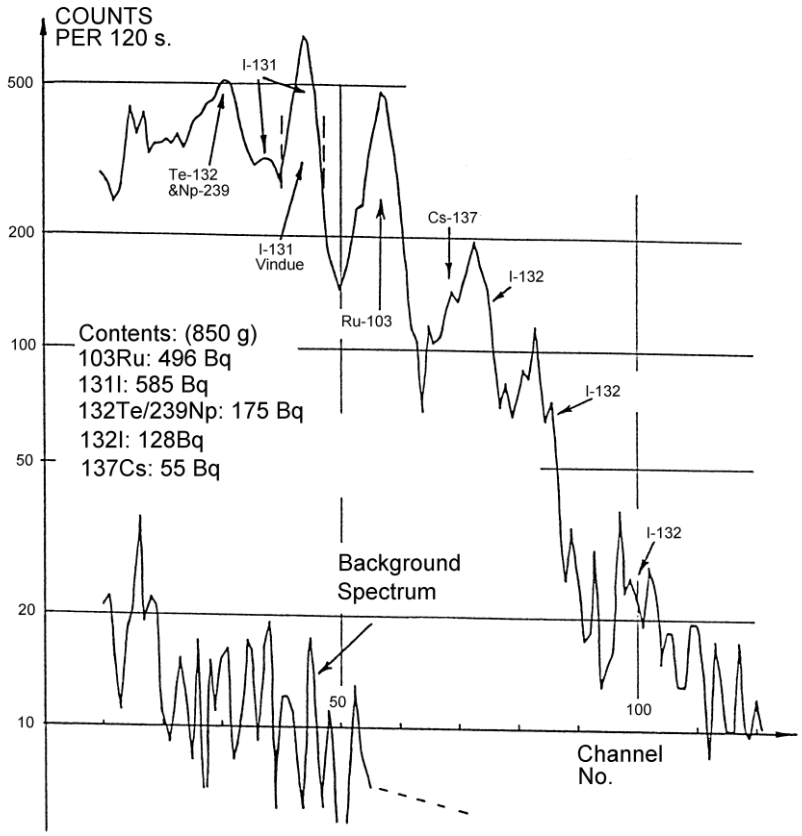

Figure 4.7: A γ-ray spectrum of rainwater collected on the 7th May 1986 in Copenhagen, measured with a 10cmx10cm NaI(Tl) detector. Taken from Hovgaard & Korsbech 1992.

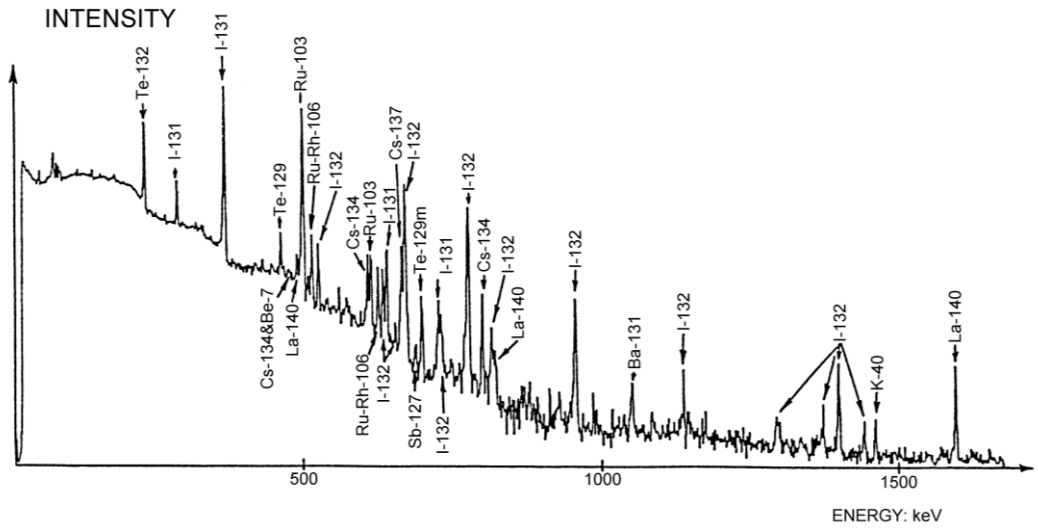

Figure 4.8: A γ-ray spectrum of rainwater collected on the 7th May 1986 in Copenhagen, measured with a Ge(Li) detector. Taken from Hovgaard & Korsbech 1992.

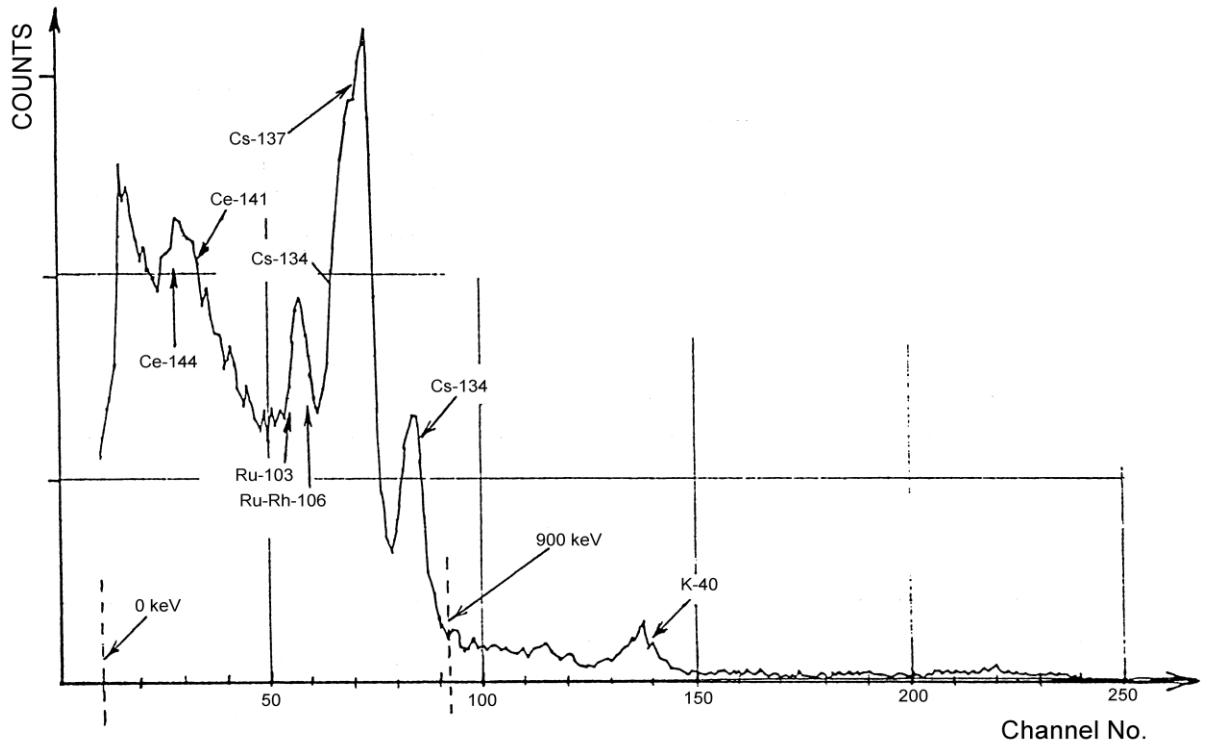

Figure 4.9: A γ-ray spectrum of mud from a gutter collected on the 13th May 1986, measured with a 10cmx10cm NaI(Tl) detector. Taken from Hovgaard & Korsbech 1992.

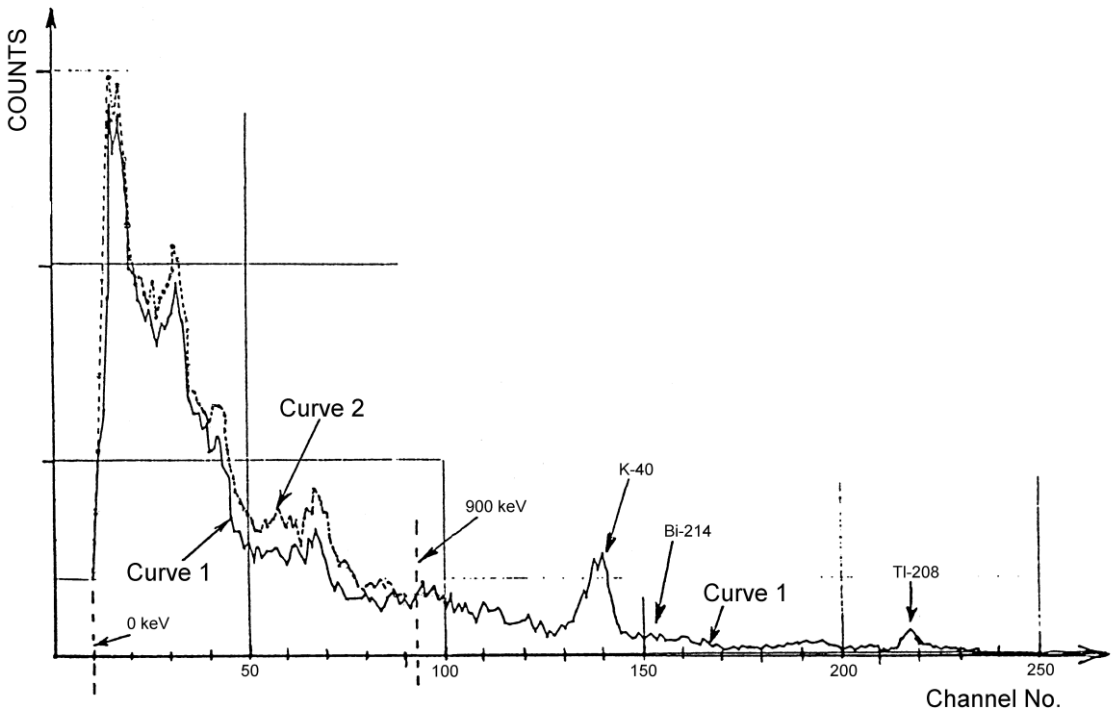

Figure 4.10: γ-ray spectra from clay samples measured with a 10cmx10cm NaI(Tl) detector. Curve 1 is for a clean sample of clay, and curve 2 is for a sample contaminated with the mud measured previously (figure 4.9). Taken from Hovgaard & Korsbech 1992.

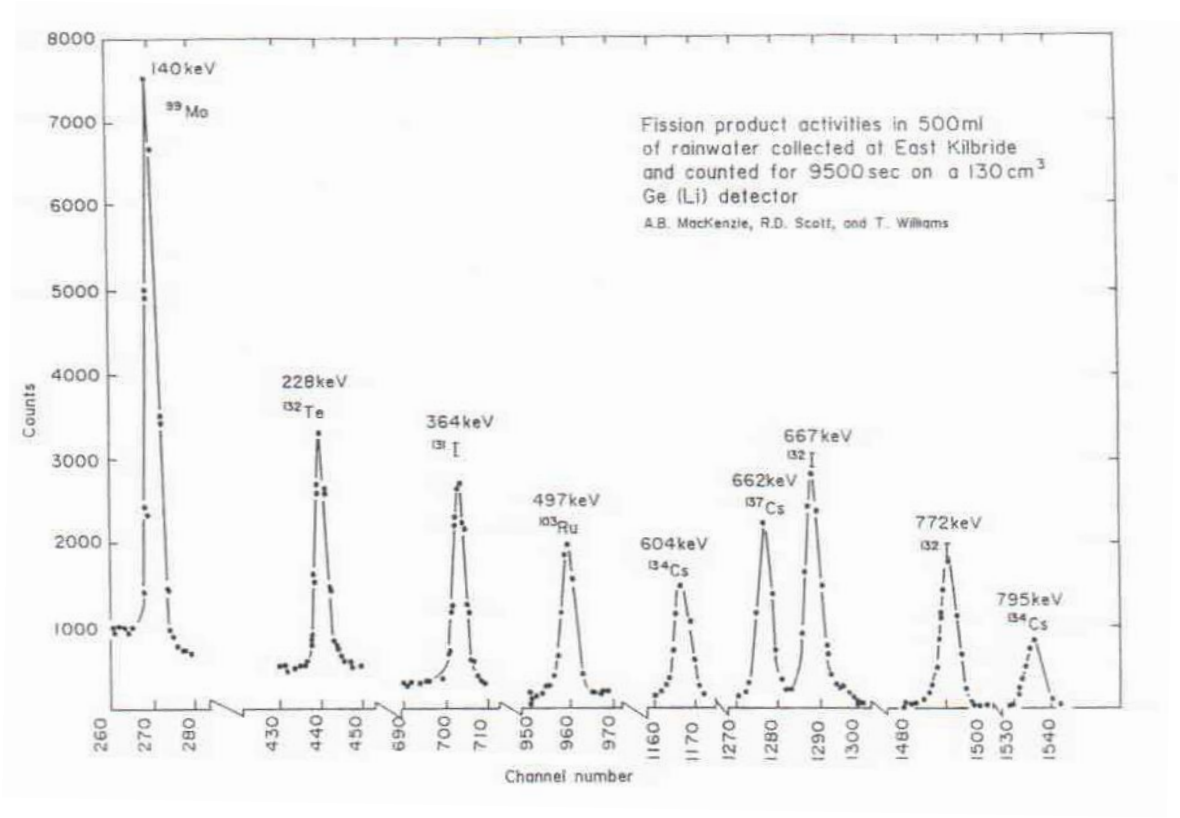

Figure 4.11: A γ-ray spectrum from rain water collected from the roof of the Scottish Universities Research and Reactor Centre on the 6th May 1986 measured with a 130 cm^3 Ge(Li) detector. The spectrum shows some of the peaks separated from the total spectrum.

Following the accident, measurements were made from clothing worn by people in the area of the accident. Figure 4.12 shows the spectrum recorded using a Ge counter on clothing worn in Kiev, taken from Ali et. al. 1986. There are several radioisotopes identifiable in this spectrum, most prominently due to ^{131}I, ^{103}Ru, $^{134,137}Cs$, ^{95}Nb, ^{95}Zr, ^{140}La and $^{141,144}Ce$.

The Chernobyl accident also resulted in the release of a large number of "hot" particles, minute particles of highly radioactive material. These particles were suspended in the air and deposited downwind of the accident, in some sites accounting for 90% of the activity (Stukin 1991). It was found that these particles could be divided into two types; those derived from fuel containing fuel material and radionuclides that had accumulated during the reactors operational life, and particles formed by the condensation of radionuclides onto an existing particle. Some particles were composed of a fuel particle onto which other radionuclides had condensed. The composition of the second type of particle varied greatly depending on the radionuclides available for condensation, the nature of the condensation nucleus and the temperature of formation. Figure 4.13 shows the spectrum from one such particle collected 1 500 m south west of the reactor, measured with a semi-conductor detector. This particle has a very strong peak due to ^{106}Ru, significantly larger than the ^{137}Cs peak, however, the spectrum recorded from a soil sample collected at the same location had a strong ^{137}Cs peak but a small ^{106}Ru peak. As would be expected, the composition of such particles can be significantly different from the bulk composition of the fall out.

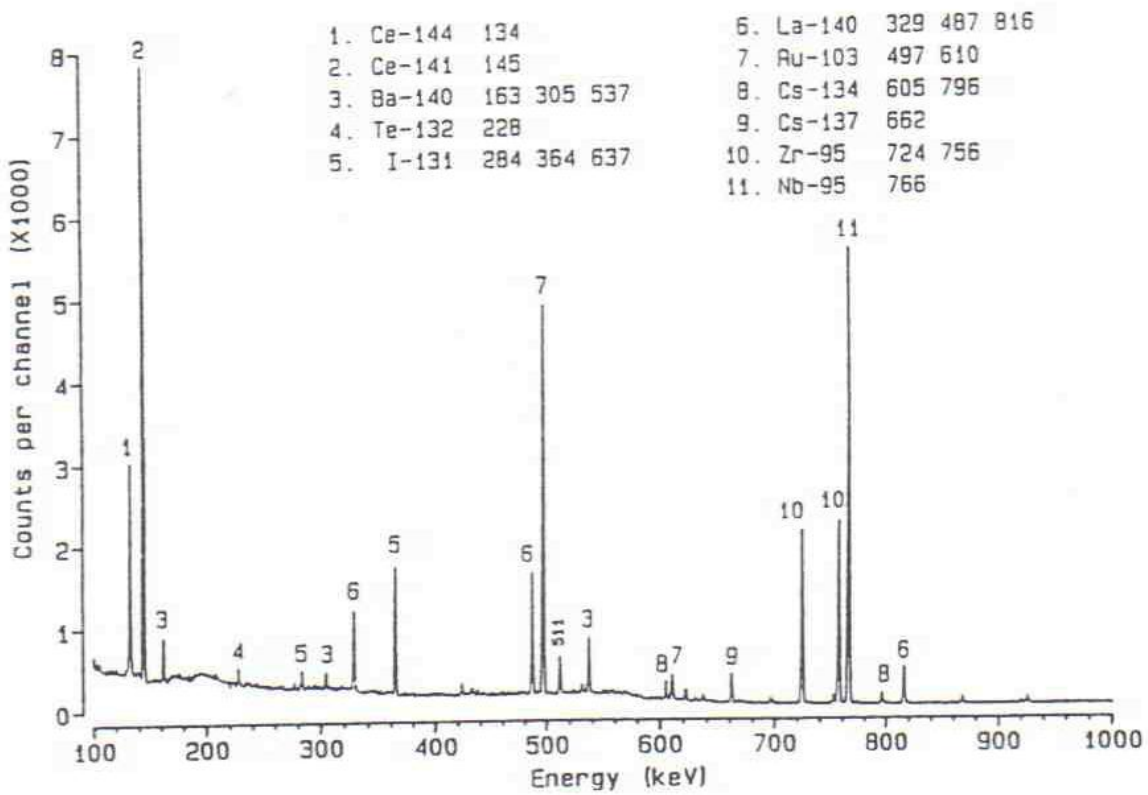

Figure 4.12: γ-ray spectrum recorded using a Ge detector from an item of clothing contaminated in Kiev. Taken from Ali et. al. 1986.

Samples taken by air filters were also studied using γ-ray spectrometers. Among these were samples collected at the German Institute for Radiation Hygiene using a high purity planar Ge detector (Winkelmann et. al. 1987) and by the AEA at Harwell, γ-ray spectra from these samples are shown in figures 4.14-4.16. These spectra are far simpler than those measured by other means, this is because air filters only measure those radioisotopes that are suspended in the air near the ground. They can not measure the radioisotopes still suspended at altitude, or those already settled on the ground. The spectrum recorded in Germany has a peak due to ^{239}Np, which was not present in later samples. The early spectrum from Harwell is dominated by Te and I isotopes, the later spectrum is dominated by Cs isotopes. The measurements made a significant time after the radioactive cloud has past are the result of resuspension of material, for example by the wind.

Whole-body monitoring was conducted on several people who had been exposed to nuclear material escaping from the reactor. Figure 4.17 shows the spectra recorded using NaI and Ge detectors from a subject who had been in Kiev for four days following the accident, taken from Ali et. al. 1986. The presence of several radioisotopes released in the accident, including iodine and caesium isotopes, are clearly visible.

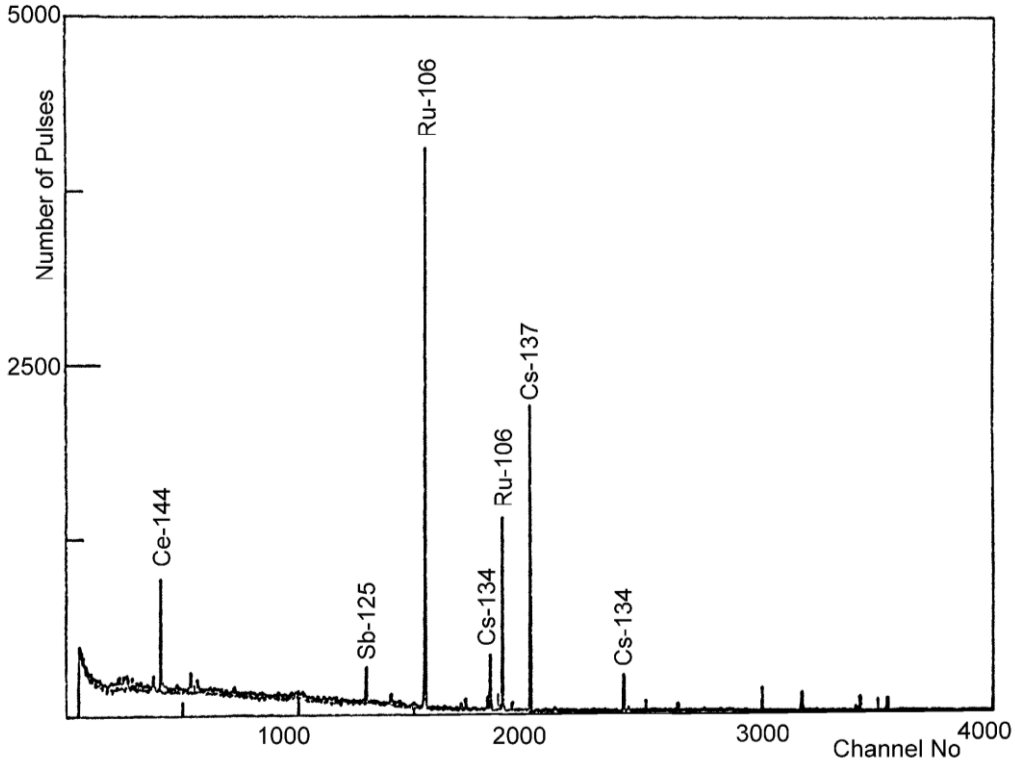

Figure 4.13: The γ-ray spectrum recorded using a Ge detector from a "hot" particle from a site 1 500 m south west of the Chernobyl reactor. Taken from Stukin 1991.

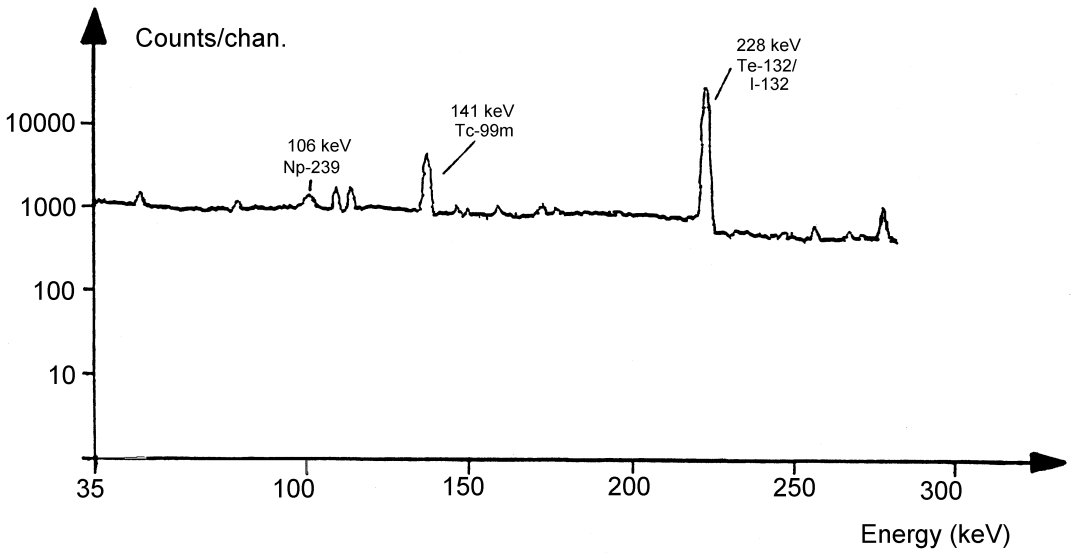

Figure 4.14: Part of a γ-ray spectrum using a high purity planar Ge detector from an air filter sample collected at the German Institute For Radiation Hygiene on the afternoon of the 1st May 1986. Taken from Winkelmann et. al. 1987.

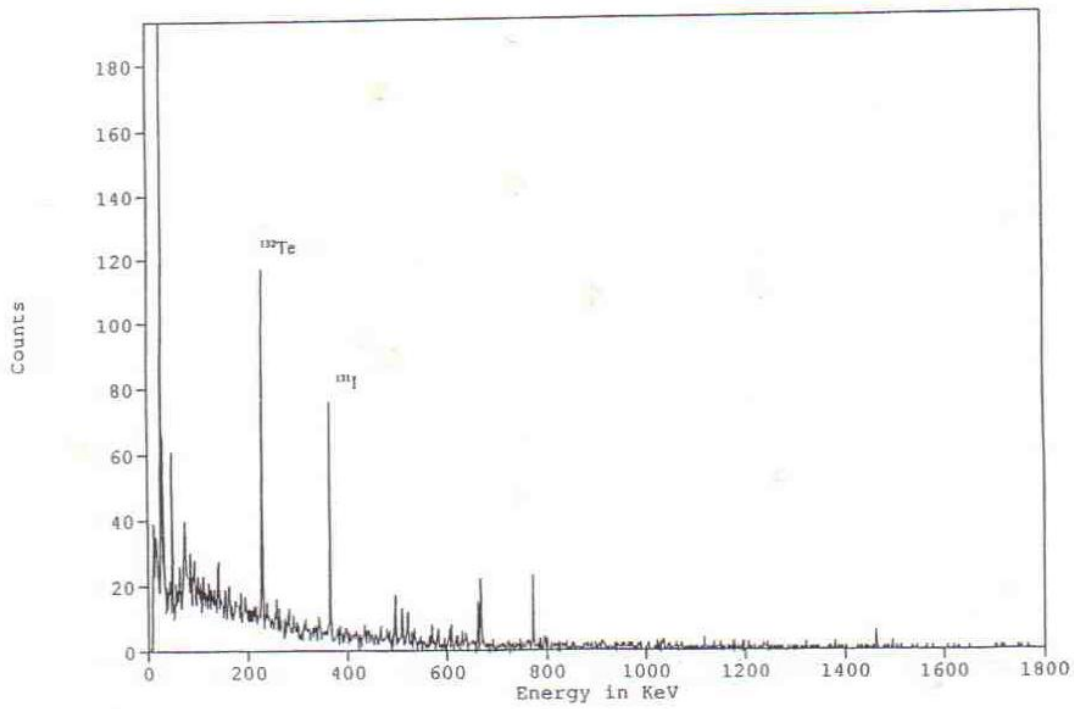

Figure 4.15: A γ-ray spectrum using a Ge detector from an air filter sample collected by the AEA at Harwell on the 2nd May 1986. Provided by Ian Adsley, AEA Gamma Spec. Lab.

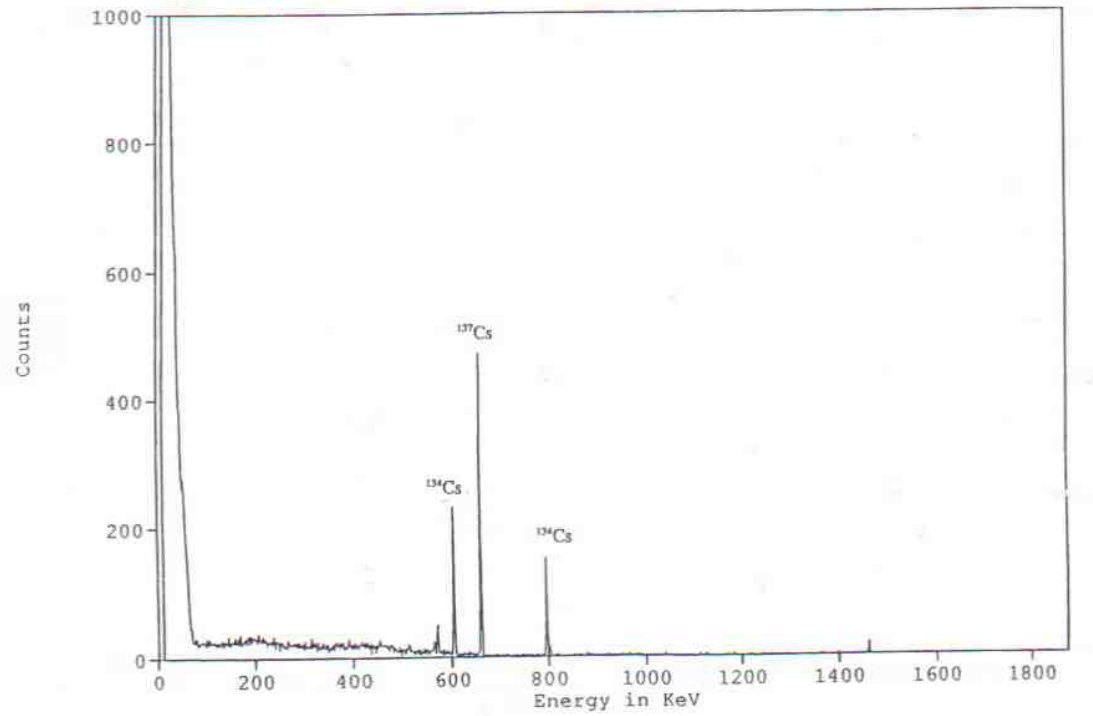

Figure 4.16: A γ-ray spectrum using a Ge detector from an air filter sample collected by the AEA at Harwell on the 2nd December 1986. Provided by Ian Adsley, AEA Gamma Spec. Lab.

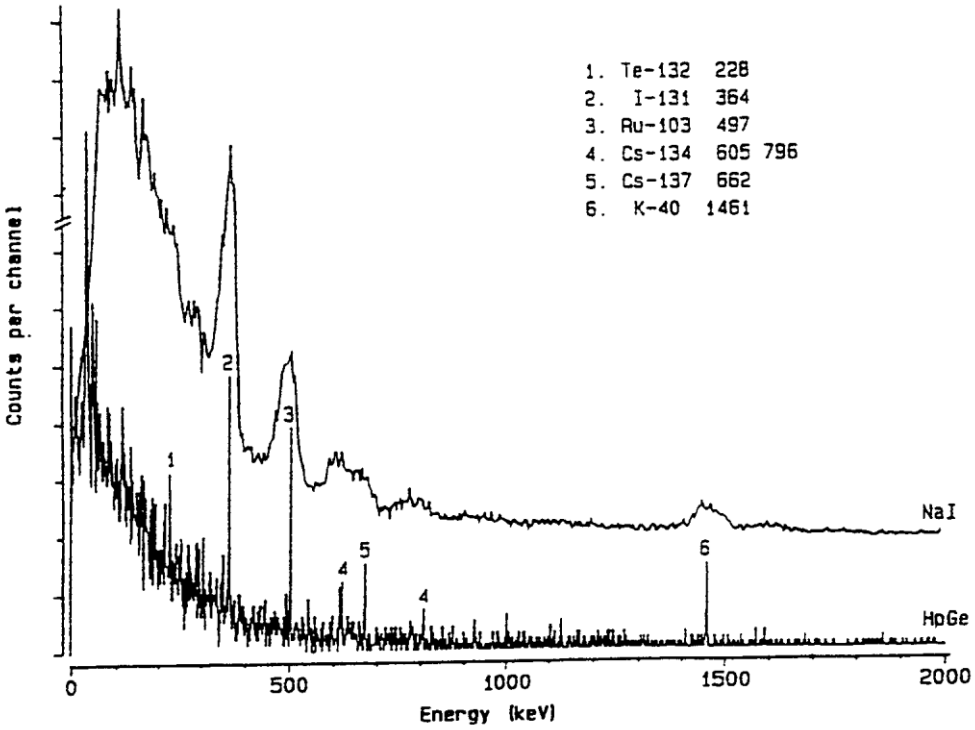

Figure 4.17: Whole body spectra of a person who had been in Kiev for four days after the nuclear accident recorded with NaI and Ge detectors. Taken fron Ali et. al. 1986.

4.3 General Features of Gamma Spectra

A number of general observations can be made about the γ-spectra observed following nuclear accidents. Firstly, as noted, the complexity of the source term depends on the state of the reactor and its containment prior to and during the accident. Accidents occurring at full power with 'old' fuel will tend to involve more complex source terms, containing short lived and long lived components, including activation products; by contrast the source terms associated with low power operation or recently fuelled systems will tend to be simpler. Also, as noted, the accident conditions, together with cladding and containment will influence the parts of the radioactive inventory released to the environment; its physical and chemical form playing a role in subsequent transport and deposition processes.

The spectra measured shortly after a major accident involving a complex release of radioactive material show a large number of γ-ray peaks, mostly at the low energy end of the spectrum. However, the majority of the peaks observed in these spectra are due to a relatively few radioisotopes, with a significant background due to the large number of weaker γ-emitters present.

In spectra recorded with NaI detectors, and even with higher resolution Ge detectors, many of the peaks are composites of several γ-rays, some of which are not identified. Thus, to determine the activity due to individual radioisotopes it is necessary to account for other γ-ray

lines included in the relevant peak. This is not necessarily straight forward, and in some cases may not be possible even with spectra recorded using Ge detectors.

Spectra recorded from an AGS system are also influenced by scattering processes in the air path, generating an increased low energy background. Also the requirement to have a short integration time (typically a few seconds) coupled with attenuation by the intervening atmosphere results in reduced statistics. This is a particular problem with Ge detectors because of their much lower intrinsic efficiency, although it would be less problematic in high radiation fields. An additional problem associated with Ge detectors is a reduction in resolution due to the aircraft vibration. If there are radionuclides still suspended in the air at the time of the survey then γ-rays from these airborne sources would also be detected. Contamination of the aircraft by airborne particles could also be a problem immediately following an accident. The separation of γ-rays emitted from the ground, the air and aircraft contamination would be particularly difficult. In addition, because of the large number of spectra recorded during an airborne survey the interpretation of such spectra has to be conducted automatically by computer, and in the situation of an emergency response this has to be rapid.

From experience of past accidents it is clear that airborne measurements are capable of locating and mapping areas of high activity. The differences between spectra recorded after the re-entry of the Cosmos-954 satellite over northern Canada (showing a loss of volatile components), and those recorded after Chernobyl B which apparently showed differences between Sweden and Finland B reflect the different conditions occurring in these cases. More detailed analysis of sources prepared under controlled conditions is needed to develop robust strategies for spectral analysis under accident conditions. However in the meantime the evidence of past accidents is that even the poorly resolved NaI spectra can be used, with appropriate ground-to-air calibration, for dose rate estimation and fallout mapping.

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APPENDIX A. Types of nuclear reactor accidents.

An accident involving the loss of, or a reduction in the flow of, coolant results in an increase in the temperature of the core. This can result in damage to fuel elements, which in turn releases radioactive materials into the coolant channels. A coolant leak would allow any gaseous or volatile radionuclides present in the coolant to enter the reactor building, and if the containment is poor, into the outside environment. Loss of liquid coolant also usually results in the remaining coolant boiling, which results in an increase in coolant voids which could result in an increase in activity (if the design has a positive void coefficient, see below). Various events can result in a reduction in the efficiency of the cooling system, and hence overheating of fuel elements. These include foreign objects blocking the flow of coolant and the loss of a turbine, which is a major heat-sink in a power plant.

Accidents also occur following sudden increases in reactor reactivity (sudden reductions in reactivity, although still `incidents', usually result in a power reduction and cause no serious damage). Reactivity can increase suddenly during the removal of control rods, or the insertion of moderator or new fuel. The resulting increase in power (or power excursion) is accompanied by a sudden burst of neutron and γ-radiation, which can harm or even kill anyone in the reactor room at the time, if there is inadequate biological shielding. The power increase also results in a sudden increase in the pressure of the coolant, which can cause mechanical damage to the coolant pipes or even the reactor pressure vessel. Accidents involving reactivity changes often have the most serious effects, since these accidents occur in very short time periods and can release a very large amount of energy. With such fast events the scram system may not be able to shut down the reactor before damage occurs, although well designed systems should prevent serious damage.

In some reactor designs activity can also increase as a result of voids developing in the coolant, ie: the reactor has a positive void coefficient. This occurs if the reactor is over moderated, with criticality maintained by the absorption of neutrons in the moderator. In such a situation boiling of the coolant, which acts as a moderator, creates voids and hence a reduction in moderator density. This results in an increase in thermal neutron flux, and the reactor goes supercritical. A positive feedback loop occurs, as more activity generates more heat, and further voiding. In most reactor designs the reactor is under moderated, and the coolant acts as significant part of the moderator. In this situation there is a negative void coefficient, in which boiling of the coolant results in a reduction in moderation. This reduces the thermal neutron flux, and hence the fission rate, resulting in a power reduction. This provides an inherent safety mechanism which is much faster than inserting control rods.

APPENDIX B. Incidents at nuclear facilities with no off-site consequences.

Date	Facility	Description of incident	Refs.
21/9/45 21/5/46	Los Alamos, USA critical assembly	Two accidents occurred following the mishandling of reflectors, resulting in power excursions. Two experimenters died, but the core was undamaged.	IAEA 1994
12/49	Water Boiler, USA research reactor	Two controls rods were manually lifted from the core, resulting in a power excursion. The reactor was undamaged.	IAEA 1994
18/4/52	JEMIMA, Los Alamos critical assembly	A computational error and violation of the operating procedure resulted in a power excursion. An automatic scram stopped the reaction, no damage was done.	IAEA 1994
12/12/52	NRX, Canada research reactor	A sequence of mistakes, ending with the withdrawal of control rods resulted in a power surge, with a positive void coefficient, destroying the reactor. About 10^{14}Bq of long lived fission fragments collected in the basement.	IAEA 1994 Bertini et. al.1980 Thompson and Beckerley 1964
15/3/53	Reprocessing plant, Chelyabinsk, USSR	A container of Pu was loaded twice, without the knowledge of the operators. Two operators were irradiated.	Puit 1996
4/1/55	Hanford KW Reactor, Washington, USA	A coolant tube was blocked by a neoprene disk, resulting in a ruptured fuel element. The entire graphite channel had to be removed through a hole cut in the rear concrete shield wall after all standard methods to remove it had failed.	Bertini et. al.1980 Thompson and Beckerley 1964
29/11/55	EBR-1, Idaho, USA research reactor	A study of fast power rise was to be terminated with a manual shut off button. Due to operator error, the wrong button was pressed. The power excursion melted 40% of the fuel elements before the correct button could be pressed..	IAEA 1994 Bertini et. al.1980 Thompson and Beckerley 1964
23/7/56	Materials Testing Reactor, Idaho, USA	During scheduled refuelling a highly active reactor component was placed in an inadequately shielded position.	IAEA 1994
21/4/57	Reprocessing plant, Chelyabinsk, USSR	U oxide precipitation accumulated in a tank. One operator died, and a further 5 were seriously irradiated.	Puit 1996
2/1/58	Reprocessing plant, Chelyabinsk, USSR	A container was tipped for emptying, the resulting change in geometry caused the solution to go critical. Three operators died, and another was blinded.	Puit 1996
23/5/58	NRU, Chalk River, Canada research reactor	Water seeped into a fuel element, causing it to burst. The building was badly contaminated while removing the damaged element from the core.	IAEA 1994 Bertini et. al.1980 Thompson and Beckerley 1964
16/6/58	Y12 reprocessing plant, Oak Ridge, USA	8 operators irradiated in a criticality accident.	Puit 1996
15/10/58	Vinca, Yugoslavia critical assembly	The reactor went supercritical following the failure of instrumentation. One person died, and another five suffered severe radiation sickness. The reactor was undamaged.	IAEA 1994
18/11/58	HTRE-3, Idaho research reactor	Faults in the design of detectors resulted in an unnoticed power increase. An automatic scram occurred, but only after several fuel elements had partially melted.	IAEA 1994 Bertini et. al.1980 Thompson and Beckerley 1964 Stratton 1967
30/12/58	Los Alamos, reprocessing plant	Pu storage limits in a tank were exceeded, which went critical killing one operator.	Puit 1996
15/3/60	Alize, Saclay, France critical assembly	Operator error resulted in the removal of a control rod, creating a power excursion. The core was undamaged.	IAEA 1994

5/12/60	Reprocessing plant, Chelyabinsk, USSR	Pu limit was exceeded during a transfer to a storage tank. Several operators were slightly irradiated.	Puit 1996
25/1/61	Reprocessing plant, Idaho, USA	After a release of compressed gas enriched U solution entered the evaporator. The area was highly contaminated.	Puit 1996
4/7/61	Submarine reactor accident	The accident occurred in the first Soviet missile-carrying nuclear submarine, and details were not released until 1990. There was a loss of pressure in the primary coolant circuit in one of the two reactors on board. The emergency shut down system functioned correctly, but a fire broke out in the reactor compartment and the submarine surfaced. Normal cooling of the reactor was not possible, and the core temperature began to rise. A temporary emergency cooling system was rigged using piping from several weapons systems and the submarines fresh water reserve, this involved welding work inside the reactor compartment. All the compartments of the submarine were contaminated, and 8 crewmen died within one month.	Kuznetzov 1991
14/8/61	Reprocessing plant, Tomsk, USSR	U accumulated in the oil tank of a transfer pump. One operator was irradiated.	Puit 1996
16/10/61	Reprocessing plant, Idaho, USA	Enriched U solution was transferred to a tank already containing waste material, which overflowed into the shielded basement.	Puit 1996
26/10/61	SPERT-3, Idaho, USA	A crack in the pressurizer was discovered along a welded seam.	Bertini et. al.1980 Heffner et. al.1962
12/12/61	Engineering Test Reactor, USA	Water flow was blocked by a plastic "sight box" used during maintenance. Melting occurred in small portions of 6 fuel elements, and the building was badly contaminated	IAEA 1994
7/4/62	Pu processing plant, Hanford, WA, USA	Pu vapours were released from a can, which should have been empty. 3 operators were irradiated over a 37hr period.	Puit 1996
7/9/62	Reprocessing plant, Chelyabinsk, USSR	Pu limit exceeded following the arrival of waste abnormally rich in Pu.	Puit 1996
7/10/62	Portable Medium Power Plant 3A (PM-3A), McMurdo Sound, Antarctica	Accumulation of hydrogen in the upper portion of the containment vessel was ignited by a short circuit (which also triggered a scram). The result was a small fire of short duration, damaging some control systems.	Bertini et. al.1980 ORNL,NSIC 1966 Buchanan 1963b
13/11/62	Materials Testing Reactor, Idaho, USA	Water flow to several fuel plates was blocked by rubber debris from a gasket. Partial melting occurred.	IAEA 1994
30/1/63	Reprocessing plant, Tomsk, USSR	For a 10hr period there was no control of the U concentration of solutions transferred to a storage reservoir.	Puit 1996
1/7/63	Oak Ridge Reactor, Oak Ridge, USA research reactor	A large neoprene gasket lodged on one of the fuel elements, blocking coolant flow. One of the fuel plates of this element was partially molten, about $4x10^{13}$ Bq of fission products entered the cooling water.	IAEA 1994 Bertini et. al.1980 Colomb and Tims 1963/4 ORNL,NSIC 1966
Autumn 1963	University of Michigan Reactor, USA	A thimble was ruptured allowing water from the pool to leak onto the experimental floor. No damage occurred.	IAEA 1994
18/10/63	University of Virginia Reactor, USA	A break in piping occurred lowering water level in the pool. No damage resulted from the loss of water.	IAEA 1994
11/63	Agriculture and Mechanics College Reactor, Texas	A leaking gasket allowed water to leak from the pool. Pool water activity was $1.6H10^{6}$ Bq ml^{-1}. No serious consequences resulted from the accident.	IAEA 1994
3/12/63	Reprocessing plant, Tomsk, USSR	For an 18hr period there was no control of the U concentration of solutions transferred to a storage reservoir.	Puit 1996
1964-79	Byeloyarsk NPP, USSR	Over a 15 year period fuel assemblies were persistently damaged by overheating. During repair work staff were over exposed.	Medvedev 1989

29/9/65	Plutonium Recycle Test Reactor (PRTR), Richmond, Washington	A rupture in a metal sheath within a coolant channel resulted in a low pressure signal and a scram. There was a release of radioactive water and gas to the containment vessel.	Bertini et. al.1980 USAEC 1965/6, 1975
13/11/65	Elektrostal fuel fabrication plant, Moscow, USSR	UO_2 powder was placed in a water tank, where it went critical.	Puit 1996
15/12/65	Reprocessing plant, Chelyabinsk, USSR	Because of an error in calculating the U content of waste the U mass limit was exceeded.	Puit 1996
30/12/65	Venus (Mol), Belgium critical assembly	During the manual removal of a control rod, the reactor became critical. The technician needed to have a foot amputated. The reactor was undamaged.	IAEA 1994
1966-75	BR2, Mol, Belgium research reactor	Over a nine year period there were 11 accidents. There were 4 leakages of fuel capsules outside the core; a 5.7 ton container fell close to spent fuel storage; a TeO_2 capsule burnt releasing $4x10^{11}$Bq of ^{131}I into reactor building; hot cells were dropped 6m when the cable of a crane fractured; fuel element cladding failed; a 20 ton container used to carry irradiated fuel fell after the fracture of two cables; and a 350 kg sodium loop fell following the fracture of a nylon cord; a screwdriver which had fallen into the core during shutdown blocked the coolant flow through one fuel element which partially melted. All incidents produced no contamination beyond the building.	IAEA 1994
7/5/66	Melekess NPP, USSR	A prompt neutron surge resulted in some exposure of staff. The reactor was extinguished when two bags of boric acid were dumped in it.	Medvedev 1989
5/10/66	Enrico Fermi Reactor, Detroit, USA	Segments of zirconium sheets added to the coolant flow guide and vessel penetration barrier tore loose, blocking coolant flow through some fuel assemblies. The bulk of the fuel in two assemblies melted.	Medvedev 1989 Bertini et. al.1980 Scott 1971
7/2/68	San Onofre, California	A fire started in electrical cabling, the reactor was safely shut down. Incorrect wiring during repairs meant a control rod remained inserted. A second fire in March resulted in the loss of power to several systems, causing some problems during shut down.	Bertini et. al.1980
15/10/68	ISIS, Saclay, France research reactor	An experimental rig with negative reactivity was removed, leading to a power excursion. A few fuel elements were damaged, there was no external radiation hazard.	IAEA 1994
10/12/68	Reprocessing plant, Chelyabinsk, USSR	The emptying of a reservoir in violation of regulations allowed Pu to accumulate. One operator died, another had his leg amputated.	Puit 1996
21/1/69	Lucens Experimental Nuclear Power Station, Switzerland	A breach in a seal of the carbon dioxide cooling system led to a complete loss of coolant pressure, and a reduction in the cooling potential of the gas. The fuel heated, and one element and cladding melted. Radioactive material, mostly ^{88}Kr, escaped to the underground cavern housing the reactor.	Bertini et. al.1980 Miller 1975 Winiger and Halter 1977
17/10/69	Saint Larent, France	During refuelling (with the reactor running) the operator over-rode automatic stops on the fuelling rig, resulting in a coolant flow restrictor rather than a fuel element being inserted. Some fuel elements melted.	Bertini et. al.1980 Corbett 1971
mid 1970	H.R. Robinson NPP, South Carolina, USA	During pre-start up pressure testing a valve burst. Seven men were injured.	Bertini et. al.1980 Casto et. al.1970 ORNL,NSIC 1972
15/5/70	La Crosse NPP, Wisconsin, USA	A malfunction caused a valve to close, stopping steam supply to the turbine. Pressure in the reactor built up and a scram occurred. Normal pressure control was upset by a faulty relief valve, the water level fell below the top of the core.	Bertini et. al.1980 Casto et. al.1970
5/6/70	Dresden-2 NPP, Illinois, USA	A spurious signal opened the valves of the turbine steam supply, causing a reactor scram. As the water level in the reactor fell this valve closed, and water level began to rise. However, the indicator in the control room stuck, and operator added more water. The reactor was flooded, the drywell contaminated and some electrics shorted.	Bertini et. al.1980 Cagle 1971

24/8/70	Windscale fuel reprocessing plant	Pu accumulated in a hydraulic system, mildly contaminating the area and irradiating two operators.	Puit 1996
30/9/70	N Reactor, Hanford, Washington, USA	A pump filter became clogged with foreign material. The primary scram system failed to activate, the reactor was shut down by the auxiliary scram system.	Bertini et. al.1980 Gallagher 1971
14/3/71	Robinson-2 NPP, South Carolina, USA	The emergency back-up battery was drained after an emergency pump was left running after a test. The reactor later scrammed, but the low battery power caused several auxiliary circuits to fail. The bearings in all 8 turbines and generators were damaged.	Bertini et. al.1980 ORNL,NSIC 1975 Casto et. al.1971
Nov 1971	Indian Point NPP, New York, USA	During construction of unit 2 a store room caught fire. The control room of unit 1 was filled with smoke, requiring breathing apparatus.	Bertini et. al.1980 Scott et. al.1972
1972	Turkey Point-3, Florida, USA	In pre-fuelling tests 3 of 4 safety valves suddenly separated from 2 headers of the main steam line in the secondary system.	Bertini et. al.1980 Casto et. al.1972
Mar 1972	Oconee-1 NPP, South Carolina, USA	21 out of 52 in-core instrument nozzles broke off, causing extensive damage to the coolant system. This was revealed before start-up.	Bertini et. al.1980 Casto et. al.1972
9/6/72	Quad Cities NPP, Illinois, USA	The reactor was shut down after river water (used to condense steam) flooded the basement.	Bertini et. al.1980 Casto et. al.1972
27/7/72	Surry-1 NPP, Virginia, USA	During regular maintenance two men died as a result of steam burns.	Bertini et. al.1980 Casto et. al.1973
1/9/72	Millstone-1 NPP, Connecticut, USA	Seawater (used to condense steam) entered the primary coolant systems as a result of corrosion of tubes in the condenser.	Bertini et. al.1980 Casto et. al.1973a,b ORNL,NSIC 1975
7/11/73	Vermont Yankee, Vermont, USA	An interlock system to prevent the simultaneous removal of more than one control rod was nullified during a test to ensure the rods were free to move. Two rods were removed at once and the reactor went critical.	Bertini et. al.1980 ORNL,NSIC 1975
Nov 1973	Surry-1 NPP, Virginia, USA	The main reactor coolant pump fractured resulting in a complete loss of coolant flow, the reactor scrammed.	Bertini et. al.1980 ORNL,NSIC 1975
10/12/73	Surry-2 NPP, Virginia, USA	An operator was seriously injured when he was sucked through an airlock door into the containment building.	Bertini et. al.1980 Casto et. al.1974
20/8/74	Beznau-1 NPP, Switzerland	A turbine shut down a result of power grid fluctuations. The failure of a bypass valve caused an increase in the pressure in the steam generator, causing a reactor scram. The relief valve opened, but failed to close and steam flowed into the drain tank. A seal in the drain tank failed allowing primary water to flow onto the floor of the containment building.	Bertini et. al.1980 Lafaille et. al.1974
22/3/75	Browns Ferry, Alabama, USA	Both reactors were shut down after polyurethane foam was ignited by a candle used to check for air leaks. The loss of some wiring complicated the supply of coolant to unit 1.	Bertini et. al.1980 Scott 1976
7/75	EWA, Swiek, Poland research reactor	A programme of radiographic testing found several defects in the welding of joints in the reactor tank.	IAEA 1994
May 1975	Robinson-2 , South Carolina, USA	About 135000 gallons of primary coolant spilt into the containment building after a seal in the primary pump broke. Escape of radioactive material to the atmosphere was contained within prescribed limits.	Bertini et. al.1980 ORNL,NSIC 1978 Casto et. al.1975
5/11/75	Cooper NPP, Nebraska, USA	Incorrect wiring indicated that a closed valve was open. This allowed hydrogen gas to accumulate in a sump, the resultant explosion injured two men.	Bertini et. al.1980 Casto et. al.1976 ORNL,NSIC 1978
7/1/76	Cooper NPP, Nebraska, USA	An ice plug inside the off gas stack resulted in a back pressure that allowed hydrogen into the off-gas building. An explosion demolished the building. Some radioactive materials were released into the vicinity of the building, but nothing beyond the site boundary.	Bertini et. al.1980 Casto et. al.1976 ORNL,NSIC 1978

late 1976	Millstone-1 NPP, Connecticut, USA	The reactor went critical and was automatically scrammed when during a safety test the wrong control rod was withdrawn.	Bertini et. al.1980 ORNL,NSIC 1978
1977	Byeloyarsk NPP, USSR	Half the fuel assemblies in the core melted. Repairs, during which the staff were exposed to radiation, lasted about a year.	Medvedev 1989
20/3/77	Rancho Seco-1, California, USA	A short circuit on the control panel (caused while an operator was replacing a bulb) cut power to two thirds of the non-nuclear instruments. The resulting erroneous signals caused the integrated control system (ICS) to shut off feedwater supply to the steam generators. The increase in primary water temperature and pressure resulted in a reactor scram. ICS kept valves closed because of erroneous signals, the temperature in the primary system was maintained by injecting cool water and correct operation of the relief valve.	Bertini et. al.1980 Casto et. al.1978
15/7/77	Browns Ferry 3, Alabama, USA	A fire occurred in a charcoal filter bed after a hydrogen build up due to a blocked drain in the air system reducing the efficiency of the catalytic recombination of hydrogen and oxygen.	Bertini et. al.1980 Mays 1979
24/9/77	Davis Besse 1 NPP, Ohio,USA	A valve controlling water supply to the steam generator closed, causing the water level in the steam generator to drop, and hence the temperature and pressure of the primary circuit to rise. The pressure relief valve stuck open, and steam escaped to the quench tank, and from there into the containment building when a rupture disk blew.	Bertini et. al.1980 Casto et. al.1978
3/3/78	Crystal River 3 NPP, Florida, USA	Parts of a burnable-poison-rod assembly were found in the steam generator, core and pressure vessel. A series of power tilt alarms (12/12/77 to 17/2/78) may have been due to the early stages of the disintegration of this assembly.	Bertini et. al.1980 Casto et. al.1979
24/7/78	Wood River Junction fuel fabrication plant, USA	As a result of procedural errors, a canister of enriched U solution was emptied into a storage tank, which went critical. One operator died, and 2 more were irradiated.	Puit 1996
17/10/78	Reprocessing plant, Idaho, USA	U accumulated in a column as a result of an undetected flow of material. The area was highly contaminated.	Puit 1996
13/12/78	Pu storage facility, Tomsk, USSR	A container for 2 ingots of Pu was loaded with 3. One operator was irradiated.	Puit 1996
31/12/78	Byeloyarsk NPP, USSR	A fire was started when a roof panel fell onto a fuel tank. The control cables burned out, and the reactor went out of control. Eight people were exposed to severe doses of radiation in an effort to supply emergency cooling water.	Medvedev 1989
1979	Montpelier	One person had a leg amputated after handling a γ-source removed from the apparatus	Pasquier 1996
2/5/79	Oyster Creek 1 NPP, New Jersey, USA	A spurious electrical signal resulted in a scram and the disconnection of recirculation pumps. Lack of power caused problems in cooling the core.	Bertini et. al.1980
13/3/80	Saint-Laurent A NPP, France	A metal plate blocked the flow of gas coolant through six channels, causing the fuel elements in these channels to melt.	Saur 1996
10/82	Armyanskaya NPP, USSR	An explosion in a generator resulted in a fire which burned down the turbine hall. The core was undamaged.	Medvedev 1989
23/9/83	RA-2, Argentina critical assembly	To save time the lone operator bypassed the safety system while changing the core configuration. A power excursion occurred which killed the operator, but the core was undamaged.	IAEA 1994
28/2/85	Virgil C. Summer NPP, S.Carolina	The reactor became critical too soon, resulting in a power surge. There was no contamination off site.	Medvedev 1989
27/6/85	Balakovo NPP, USSR	During start-up a relief valve burst filling a room where people were working with steam at 300EC. 14 people were killed.	Medvedev 1989

1/5/86	Agricultural and Mechanical University, Texas	An experimental device with negative activity was withdrawn from the reactor resulting in a power spike. No damage occurred.	IAEA 1994
12/1/87	Saint-Laurent A NPP, France	Large chunks of ice blocked the water inlet for the secondary coolant. This caused the condensers for the heat exchangers to over pressurize and caused the reactor to automatically shut down.	Leblond 1996
29/2/88	North Carolina State University, USA	A leak in the primary coolant system kept the reactor shut down, release limits were not exceeded.	IAEA 1994
10/10/88	EWA, Poland research reactor	A fuel element was dropped and broke. About $2x10^9$ Bq of radioactive material entered the reactor pool, there was no exposure to personnel.	IAEA 1994
11/11/89	TRIGA research reactor, Pitesti, Romania	During fuel shuffling two fuel assemblies were placed in free positions beyond the reach of the control rods. The reactor became critical for about 2 minutes.	IAEA 1994
20/3/90	Vogtle 1 NPP	While shut down for refuelling and routine maintenance a vehicle accident resulted in the loss of offsite AC power. Problems with the buck-up power supply resulted in a loss of shutdown cooling. Emergency power was restored before the core overheated.	Manning 1991
1991	Forbach	Two operators seriously irradiated at an electron accelerator.	Pasquier 1996
9/2/91	Mihana 2, Japan	The primary cooling circuit was breached when a tube ruptured in a steam generator.	Sandon 1996
5/4/91	High Flux Reactor, Grenoble, France	During routine inspection cracks in a water flow distributor were noticed. The component was replaced.	IAEA 1994
7/5/91	University of Massachusetts	The reactor was operated for 16 minutes with a basket, to prevent bypass flow around fuel elements, removed in violation of technical specifications. The reactor was shut down when the condition became known. No damage was done.	IAEA 1994
8/6/92	Ford Research Nuclear Reactor, Michigan, USA	A fuel element was removed while the reactor was critical, causing the reactor to shut down.	IAEA 1994
13/11/92	Iowa State University, USA	During a scram as part of an experiment it was noted that the shim rod had not fully inserted. The reactor shut down without it.	IAEA 1994
5/3/93	Ohio State University, USA	During normal shutdown it was noted that one of the safety rods failed to insert, due to a short circuit. The reactor shut down without it.	IAEA 1994
24/3/93	Ford Research Nuclear Reactor, Michigan, USA	The licensed power limit was exceed by 15% for about 11 minutes. This violated procedure, but the power level was well within the safety limit of the reactor, and below the automatic scram point.	IAEA 1994
28/4/93	University of Virginia, USA	The reactor was operated for about 5 hours with several scram systems inoperable, the problem was discovered at the end of the day when a period scram did not occur.	IAEA 1994
4/1/94	McMaster Nuclear Research Reactor, Ontario, Canada	During refuelling safety rods were withdrawn to 85% rather than the required 40%. On insertion of the sixth fuel assembly a blue glow was seen and the reactor tripped. There was no release of radioactive materials.	
Dec 1995	Monju prototype fast breeder reactor, Japan	Liquid sodium coolant leaking from pipes almost melted through the steel floor. This could have resulted in an explosive reaction between the sodium and concrete, which contains over 50% water.	