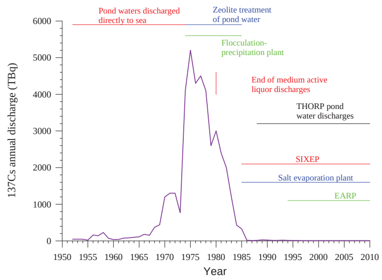
Scottish Universities Environmental Research Centre

University of Glasgow

# UK experience from the long-term assessment of fallout radionuclide mobility and operational releases

**Dr Gillian MacKinnon**
SUERC

## Timeline – key dates




Annual discharges of $^{137}$Cs from Sellafield

Dounreay

**1950's** 1954 1957 1963 1980 **1986** 2011 2013

Windscale fire

Peak weapons testing

Discharge to sea stopped

Fukushima Dai-ichi

present

**Sellafield**

**Chernobyl**

## Reactor & reprocessing operations at Sellafield

Reactor operations

- 1947 construction started on Windscale piles
- 1950 Pile 1 start up, 1952 Pile 2 start up
- 1957 Shut down
- 1956 – 2003 Calder Hall Four Magnox reactors operated
- 1963 – 1981 Windscale AGR operated

Reprocessing operations

- 1952 primary separation plant for Windscale fuel
- 1952-1954 First Pu purification plant
- 1954-1964 Second Pu purification plant
- 1963-1981 Prototype AGR fuel reprocessing plant
- 1964-1987 Integrated Magnox fuel separation & purification plant
- 1987-present New Magnox reprocessing plant
- 1994-present THORP reprocessing plant

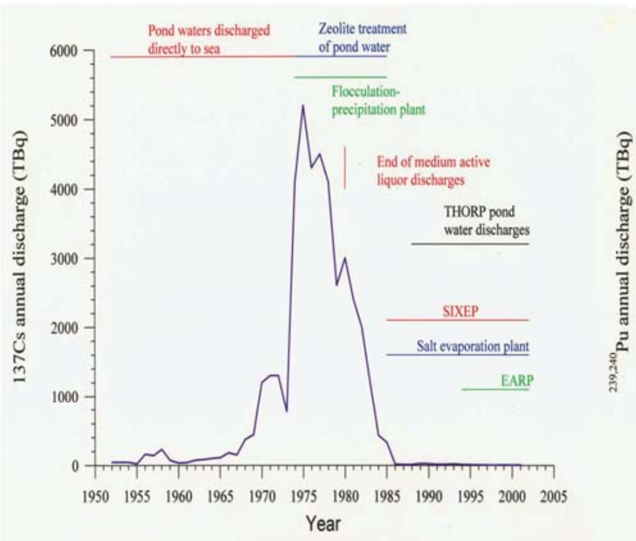
## Windscale/Sellafield site

# Effluent treatment & discharge operations at Sellafield

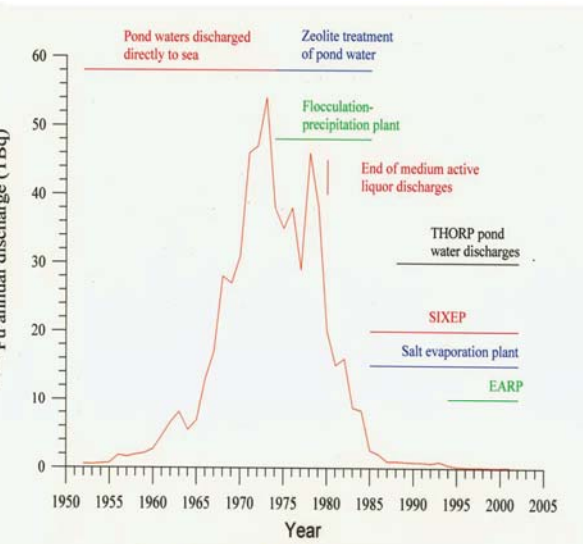
- 1952 – 1975 Pond storage water discharged directly to sea
- 1952 – 1964 Medium active waste (MAW) stored then discharged to sea
- 1964 – 1980 Evaporator used to concentrate MAW before discharging to sea
- Mid 1970's Ion exchange used for treatment of pond storage water & a new plant built for flocculation/precipitation of actinides
- 1980 Discharge of MAW stopped
- 1985 > SIXEP and new evaporator plant
- 1988 Discharges of THORP pond water started
- 1994 EARP operations started
- 2004 $^{99}Tc$ abatement technology operational

# Annual waste discharges

The changes in effluent treatment and discharge operations resulted in pronounced variations in annual discharges, with peak releases of most radionuclides in the mid 1970s as illustrated below for $^{137}Cs$ and $^{239,240}Pu$.




Annual discharges of $^{137}Cs$ from Sellafield




Annual discharges of $^{239,240}Pu$ from Sellafield

# Behaviour of Sellafield waste radionuclides

• During the period of maximum discharges, approximately 90 % of the $^{137}Cs$ remained in solution and was transported out of the Irish Sea, northwards around the Scottish coast and then into more distant waters.

Estuarine, Coastal and Shelf Science (1981) 13, 59–67

A Simple Model of Radiocaesium Transport from Windscale to the Clyde Sea Area

I. G. McKinley,* M. S. Baxter
Department of Chemistry, Glasgow University, Glasgow, G12 8QQ, U.K.

and W. Jack
Department of Natural Philosophy, Glasgow University, Glasgow, G12 8QQ, U.K.

Keywords: radioactivity; currents; Irish Sea

Introduction

Estuarine, Coastal and Shelf Science (1981) 13, 69–82

Tracer Applications of Radiocaesium in the Sea of the Hebrides

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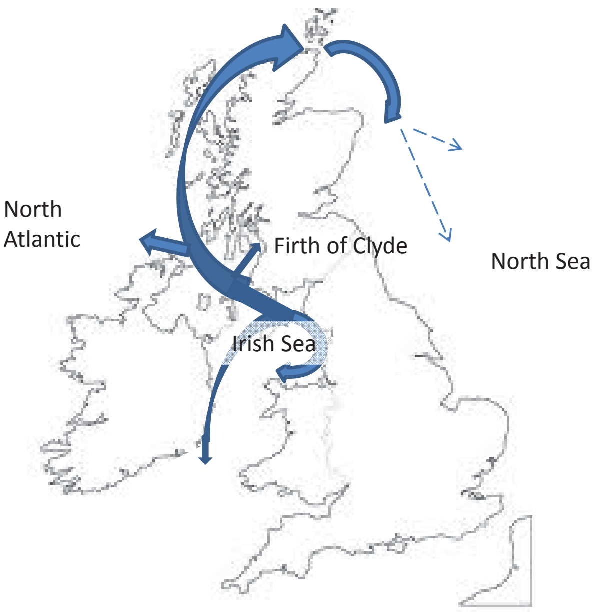
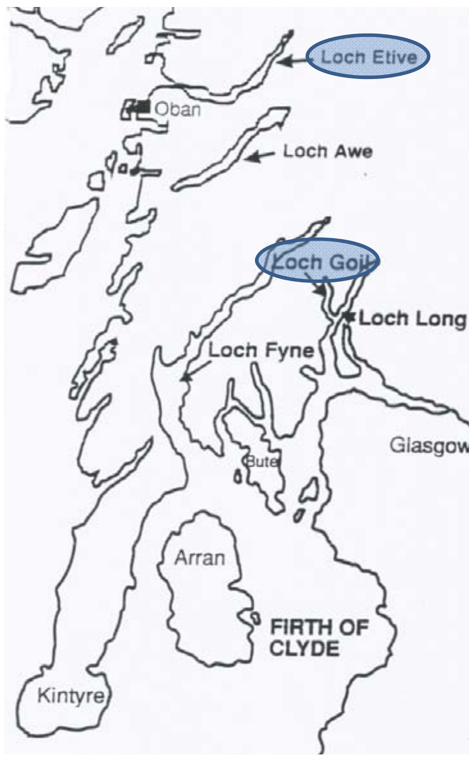
Department of Natural Philosophy, University of Glasgow, Glasgow G12 8QQ, U.K.

Introduction

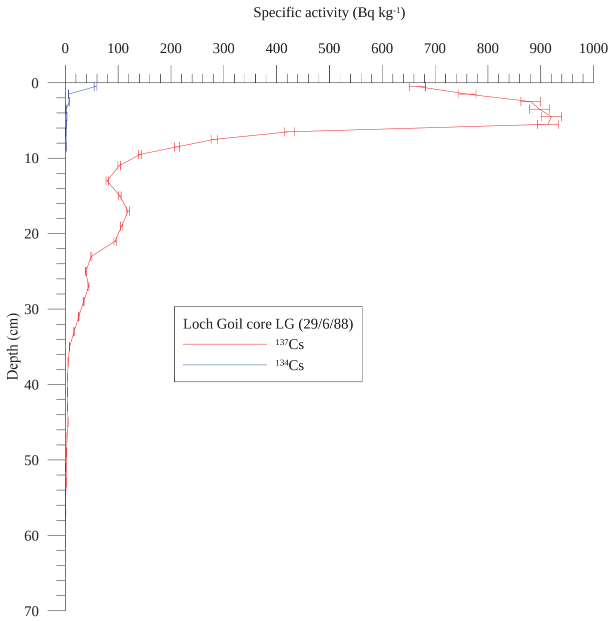

# Behaviour of Sellafield waste radionuclides

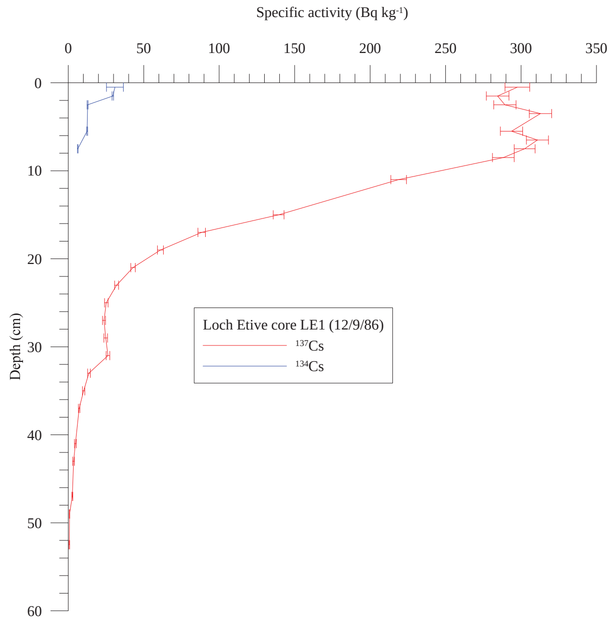



The Windscale Cs plume
(Reproduced from Jefferies et al., 1973)



# Behaviour of Sellafield waste radionuclides




Near shore environments : near conservative behaviour

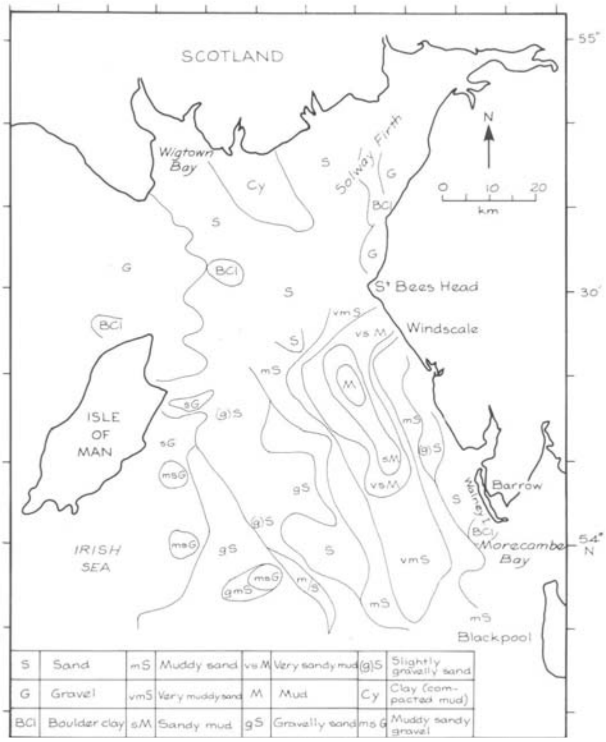
Sea lochs Goil (less intense mixing, lower accumulation rate 0.07g $cm^{-2}$ $y^{-1}$) and Etive (more intense mixing, higher accumulation rate 0.1g $cm^{-2}$ $y^{-1}$)

# Behaviour of Sellafield waste radionuclides

- During the period of maximum discharges, approximately 90 % of the $^{137}Cs$ remained in solution and was transported out of the Irish Sea, northwards around the Scottish coast and then into more distant waters.

- Approximately 10 % of the $^{137}Cs$ was incorporated in the mudpatch sediments, mainly associated with the clay component.

# Behaviour of Sellafield waste radionuclides




Surface sediment distribution according to Pantin (1978)

# Behaviour of Sellafield waste radionuclides

•Significant contamination of Irish Sea offshore sediment, with estimated inventories of 2,200 TBq of $^{137}$Cs, 549 TBq of $^{239,240}$Pu and 940 TBq of $^{241}$Am.

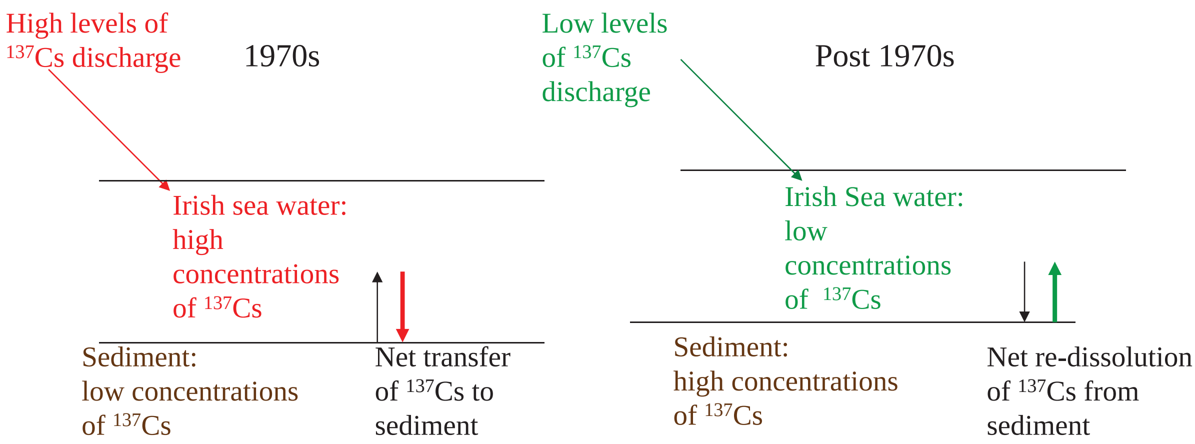
•In the late 1970s, the most highly contaminated sediment was restricted to a zone extending approximately 15 km north and 10 km south of the Sellafield pipeline and 5 km offshore.

•The contaminated sediment has subsequently been subject to gradual dispersion, with the result that variations in sediment texture now play a dominant role in governing radionuclide distributions rather than distance from Sellafield.

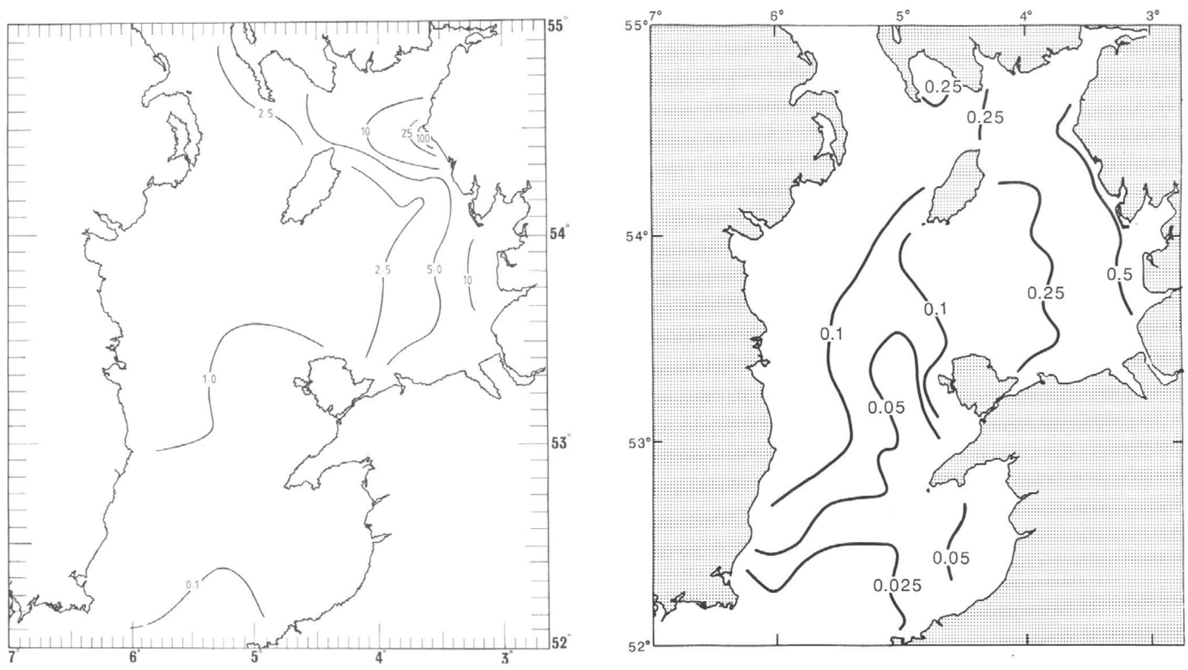
# Behaviour of Sellafield waste radionuclides

•Since the decline in discharges, operation of the law of mass action has resulted in re-dissolution of approximately 90 % of the radiocaesium from the intensively mixed top 10 cm of sediment, with less intense re-dissolution at greater depths.

• Reported $K_d$s have been of the order of $10^3$ for seawater-suspended particle systems, but of the order of $10^5$ for equilibrium between seawater and mudpatch or saltmarsh sediment.




# Behaviour of Sellafield waste radionuclides




Concentration (Bq $kg^{-1}$) of $^{137}$Cs in filtered water from the Irish Sea, 1977 and 1987.

# Behaviour of Sellafield waste radionuclides

Onshore movement of the contaminated sediment is the dominant mechanism of transport of Sellafield waste radionuclides to intertidal and saltmarsh areas of the Irish Sea.

Despite the three orders of magnitude reduction in discharges, radionuclide concentrations in sediment being deposited in these areas have not shown a proportionate reduction.

Onshore transport has been particularly important in saltmarshes, which act as sites for selective trapping and deposition of fine sediment with high radionuclide concentrations.

Cores from accreting saltmarsh deposits preserve a record of the time integrated Sellafield discharge and the radionuclide distributions can be used to derive chronologies for such deposits.

J. Environ. Radioactivity 23 (1994) 39-69

**Sediment Radionuclide Profiles: Implications for Mechanisms of Sellafield Waste Dispersal in the Irish Sea**

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(Received 5 October 1992; revised version received 1 February 1993; accepted 3 March 1993)

ABSTRACT

*Evaluation of the radiological significance of Sellafield waste radionuclide discharges to the Irish Sea requires characterisation of redistribution processes affecting contaminated marine sediment, and investigations of sediment radionuclide concentration profiles are often used in this context. Results are presented here for a study of radionuclide concentrations in saltmarsh sediments in south-west Scotland, which provide a record of Sellafield waste radionuclide deposition since the 1960s. Comparison of the results with data for the Sellafield discharge and other sediment studies indicates that the dominant mechanism of transport of Sellafield waste to the study sites involves redistribution of contaminated silt. It is also observed that the sediment profiles preserve a record of the time-integrated Sellafield discharge rather than of the annual variations. Temporal variations in sediment radionuclide concentrations are explained in terms of a simple model which includes the effects of dispersion/dilution of this contaminated silt and of radionuclide redissolution. It is concluded that, provided Sellafield discharges do not increase substantially, continuation of these processes will result in a decrease with time of pollutant radionuclide concentrations in the sediment being deposited, but in an increase in radionuclide inventories in accumulating Irish Sea intertidal and saltmarsh sediments.*

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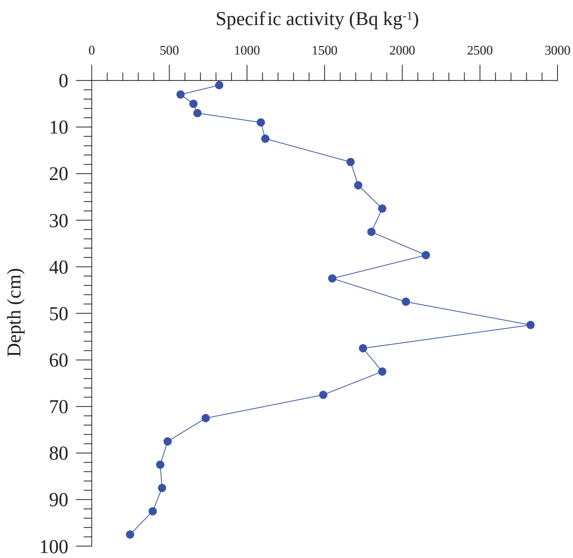
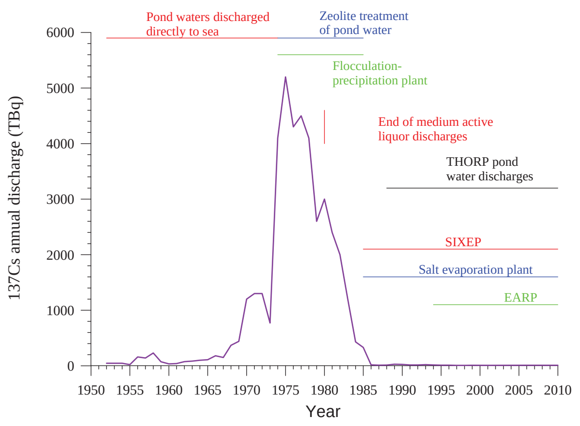
J. Environ. Radioactivity 0265-931X/94/$07.00 © 1994 Elsevier Science Limited, England. Printed in Ireland.

# Time integrated signal of Sellafield discharge

Non-conservative $^{137}Cs$ : mixed off-shore sediment deposited on-shore




Vertical distributions of $^{137}Cs$ in a Solway Firth saltmarsh core (1986)

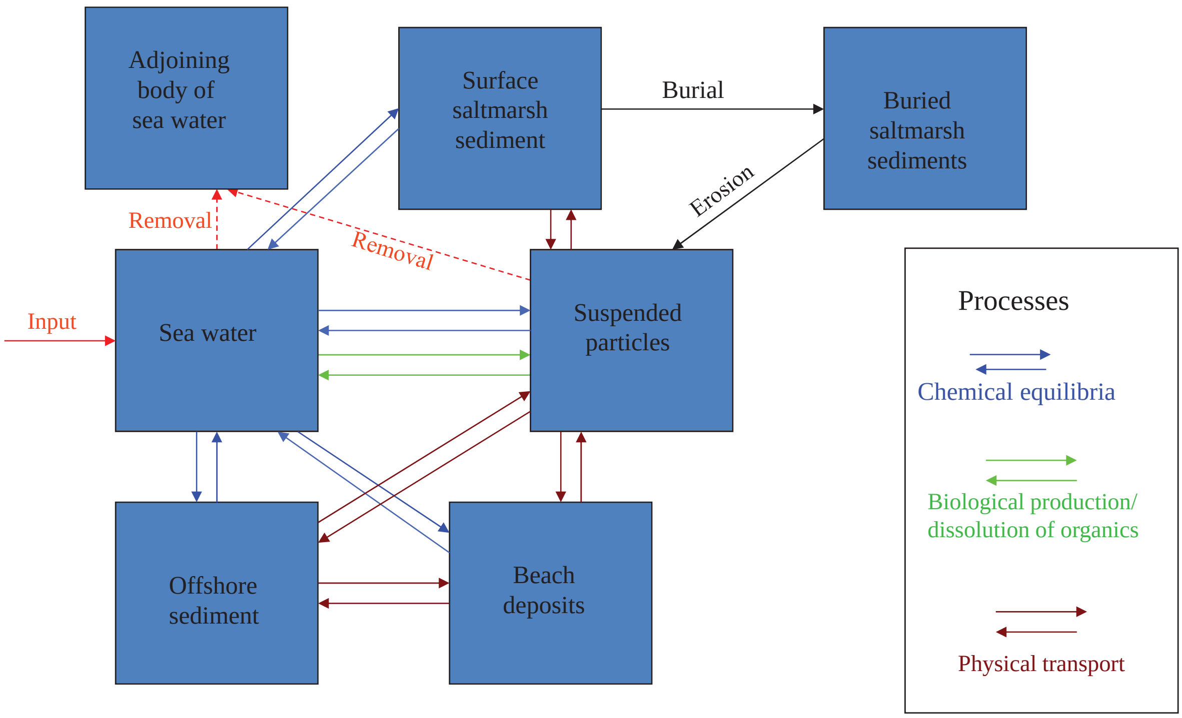



Annual discharges of $^{137}Cs$ from Sellafield

# Time integrated signal of Sellafield discharge

Wigtown Bay salt marsh

# Behaviour of Sellafield waste radionuclides




Simple box model of radionuclide behaviour in the NE Irish Sea

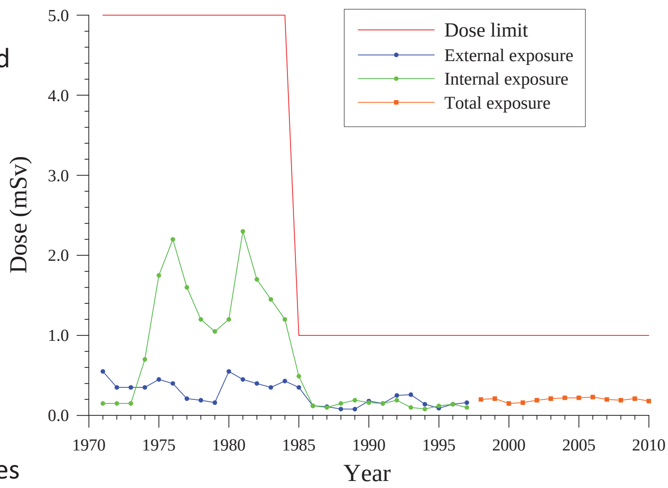
# Critical Group Exposure

The large reduction in discharges from Sellafield in the 1970s resulted in a rapid reduction of contaminant radionuclide concentrations in seawater.

However, a large inventory of radionuclides remained in the sediment, with potential for re-dissolution, transfer into the food chain and transfer to areas such as beaches and saltmarshes where contact with the human population can occur, leading to external exposure or, potentially, ingestion.

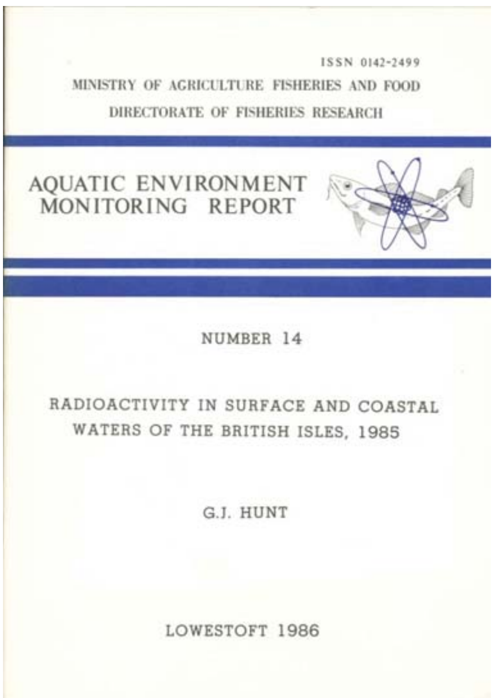
As a consequence of this environmental persistence of the contaminant radionuclides, critical group dose rates have decreased by only one order of magnitude since the period of peak releases.

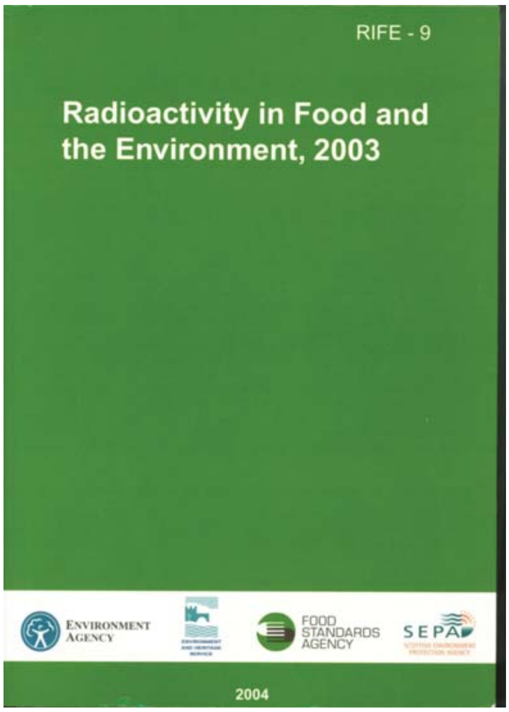
The environmental persistence of radionuclides means that the dilute and disperse approach has only been partially successful for the Sellafield discharge.




Critical group exposure from Sellafield liquid effluent discharges

# Routine Monitoring

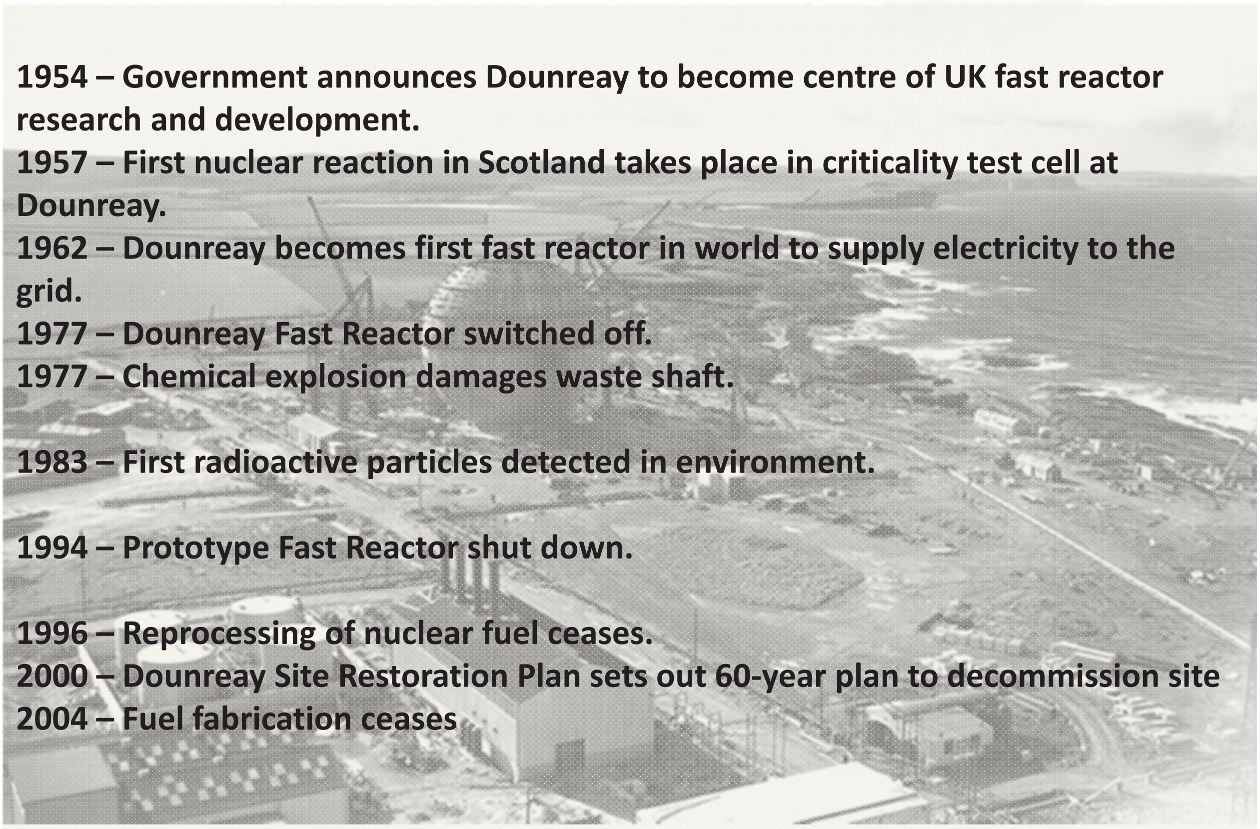



http://www.sepa.org.uk/radioactive_substances/publications/rife_reports.aspx

# Dounreay


# Dounreay 1954 - 1994



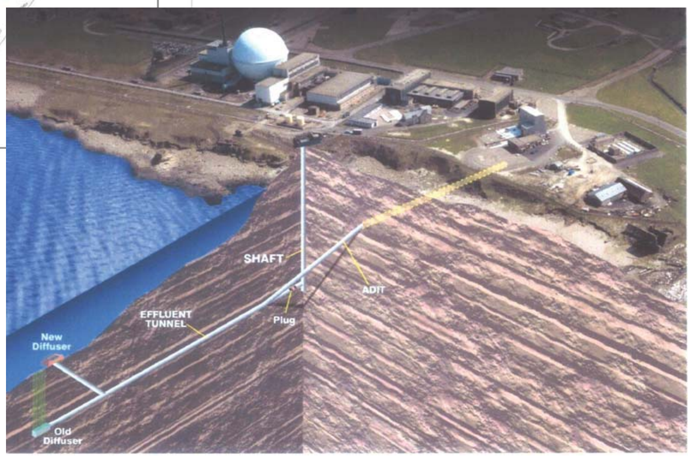
**1954 – Government announces Dounreay to become centre of UK fast reactor research and development.**
**1957 – First nuclear reaction in Scotland takes place in criticality test cell at Dounreay.**
**1962 – Dounreay becomes first fast reactor in world to supply electricity to the grid.**
**1977 – Dounreay Fast Reactor switched off.**
**1977 – Chemical explosion damages waste shaft.**

**1983 – First radioactive particles detected in environment.**

**1994 – Prototype Fast Reactor shut down.**

**1996 – Reprocessing of nuclear fuel ceases.**
**2000 – Dounreay Site Restoration Plan sets out 60-year plan to decommission site**
**2004 – Fuel fabrication ceases**

# Dounreay


The Dounreay low level liquid waste discharge pipe.

(source: Dounreay particles advisory group 4th Report, 2008)

# Dounreay

The Dounreay low level liquid waste disposal pipe tunnel and diffuser plant at the time of construction.

(source: Dounreay particles advisory group 4th Report, 2008)

# Dounreay hot particles

• Metallic, radioactive particles, consisting dominantly of MTR and PFR fuel fragments occur in the seabed and beach sediment in the vicinity of Dounreay.

• The particles are believed to have originated from historical low level waste discharges.

• It is estimated that there are thousands of particles in the seabed sediment and the waste pipeline tunnel and diffuser plant.

• Particles have also been found on the beach.

• Onshore transport is likely to represent a long term mechanism of supply of the particles to beaches, so efforts are being made to retrieve the particles from the seabed and obsolete waste disposal systems.



# Dounreay hot particles

| Year | Number of particles | Mean $^{137}Cs$ activity (MBq) | Mean depth in sediment (cm) |
|---|---|---|---|
| 1984 | 26 | 9.1 | 20 |
| 1985 | 10 | 4.9 | 10 |
| 1986 | 17 | 3.7 | 13 |
| 1987 | 10 | 9.3 | 12 |
| 1988 | 11 | 5.4 | 7 |
| 1989 | 15 | 7.7 | 8 |
| 1990 | 11 | 2.1 | 12 |
| 1991 | 13 | 2 | 16 |
| 1992 | 4 | 4 | 4 |
| 1993 | 13 | 2.7 | 12.6 |
| 1994 | 13 | 3.5 | 5.8 |
| 1995 | 11 | 5.5 | 11.5 |
| 1996 | 20 | 1.9 | 16.8 |

| Year | Number of particles | Mean $^{137}Cs$ activity (MBq) | Mean depth in sediment (cm) |
|---|---|---|---|
| 1997 | 10 | 2.1 | 9 |
| 1998 | 6 | 4.7 | 6 |
| 1999 | 11 | 3.3 | 16.6 |
| 2000 | 6 | 4.5 | 4.5 |
| 2001 | 3 | 2.7 | 2.3 |
| 2002 | 5 | 1.3 | 1.9 |
| 2003 | 3 | 2.4 | 6.7 |
| 2004 | 9 | 0.34 | 9.4 |
| 2005 | 7 | 1.4 | 8.6 |
| 2006 | 4 | 2.1 | 11.8 |
| 2007 | 9 | 2.8 | 6.7 |
| 2008 | 4 | 5.4 | 9.3 |
| | | | |

Summary of radioactive particle finds on the Dounreay foreshore 1984 - 2008

# Dounreay

# Work starts on £100m tomb for Dounreay nuclear waste

Building of storage vaults to create 100 jobs

DAVID ROSS
HIGHLAND CORRESPONDENT

WORK has started on a £100 million project to build six 65ft-deep vaults to store nuclear waste at Dounreay, each covering an area the size of a football pitch.

Around 100 jobs are expected to be created in the construction of the facility which will be the first of its type to be built in Scotland since the 1950s and the first ever to be granted planning permission.

Some 240,000 tonnes of low-level radioactive waste requires to be stored from the decommissioning of the Caithness nuclear site. The waste includes paper, rags, tools, glass, concrete, clothing and scrap metal which have small amounts of radioactivity.

By volume, this low-level waste represents more than 80% of all the radioactive waste generated by Dounreay's demolition but by radiological hazard

off and left for 300 years, to allow the radioactivity to reduce. But they will always be monitored.

The decommissioning waste is collected in 200-litre drums that are processed on site. Each

DECOMMISSIONING: low-level waste from the plant will be packed in barrels and then buried in six large vaults. Main picture: Gordon Terris

the decommissioning, said: "Cleaning out and knocking

safe disposal route for much of that waste. It is the culmination

Proposed low level waste repository at Dounreay

(source: The Herald, 26/11/11)

# Chernobyl

| Nuclide | Half life | Core inventory (EBq) | Estimated release (EBq) | % release |
|---|---|---|---|---|
| $^{85}$Kr | 10.73 | 0.033 | 0.033 | 100 |
| $^{89}$Sr | 50.5 d | 2.4 | 0.094 | 4.0 |
| $^{90}$Sr | 28.6 y | 0.20 | 0.0081 | 4.0 |
| $^{95}$Zr | 64.0 d | 5.0 | 0.16 | 3.2 |
| $^{103}$Ru | 39.4 d | 4.8 | 0.14 | 2.9 |
| $^{106}$Ru | 368 d | 2.0 | 0.050 | 2.9 |
| $^{131}$I | 8.04 d | 3.3 | 0.67 | 20 |
| $^{133}$Xe | 5.24 d | 1.7 | 1.7 | 100 |
| $^{134}$Cs | 2.06 y | 0.14 | 0.019 | 10 |
| $^{137}$Cs | 30 y | 0.28 | 0.037 | 13 |
| $^{140}$Ba | 12.8 d | 5.0 | 0.28 | 5.6 |
| $^{141}$Ce | 32.5 d | 5.6 | 0.13 | 2.3 |
| $^{144}$Ce | 284 d | 3.1 | 0.088 | 2.8 |
| $^{239}$Np | 2.36 d | 30 | 0.97 | 3.2 |
| $^{238}$Pu | 87.7 y | 0.0009 | $2.6 \times 10^{-5}$ | 3 |
| $^{239}$Pu | $2.41 \times 10^{4}$ y | 0.0009 | $2.6 \times 10^{-5}$ | 3 |
| $^{240}$Pu | $6.57 \times 10^{3}$ y | 0.0012 | $3.7 \times 10^{-5}$ | 3 |

Estimated core inventory and atmospheric release of radionuclides from the Chernobyl nuclear reactor, 1986 (source: UNSCEAR)

# Chernobyl

## Survey of radiocaesium concentrations in grassy vegetation throughout the UK

Initial Survey Aim:

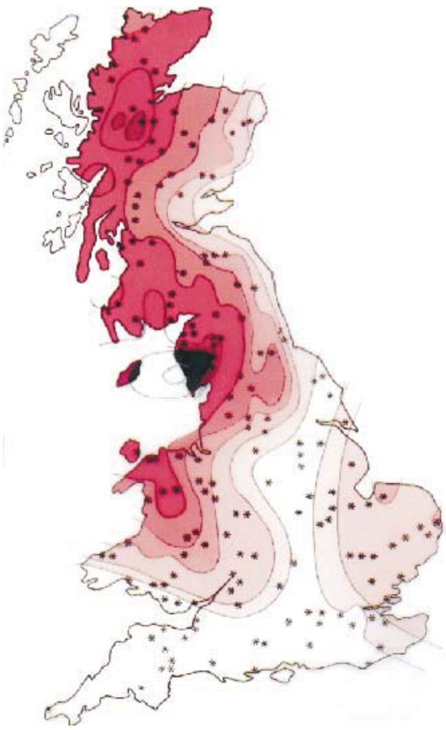
- to determine deposition patterns and levels of Cs over the country
- To establish a baseline for the movement and distribution of Cs deposited in the environment

16 land classes were sampled – 320 sampling sites – one vegetation type

Field surveyors instructed to:

- sample only grassy vegetation
- not close to roadsides
- away from overhanging vegetation

# Chernobyl



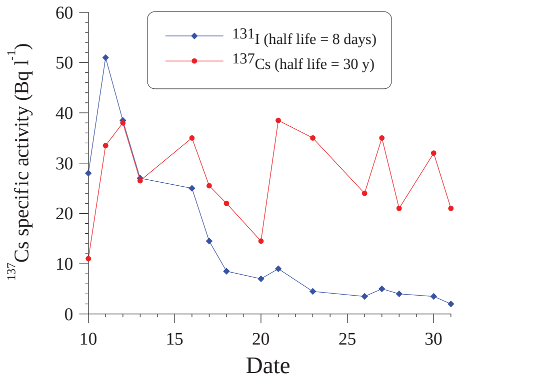
Caesium deposition on vegetation (Bq $m^{-2}$) May 1986

Allen, S.E. 1986. Radiation: a guide to a contaminated countryside. The Guardian, 17

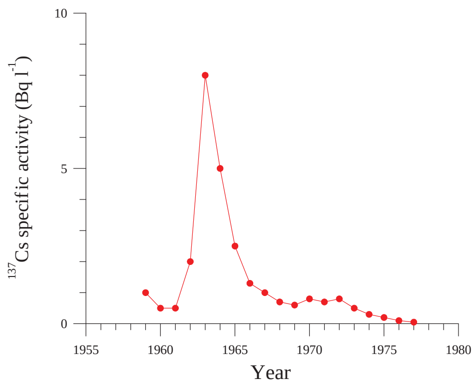
# Chernobyl

Agricultural ecosystems, in terms of radiation dose, are important at the early stages of most accidents for the majority of radionuclides as they provide the main food for most of the exposed population.

One of the most important agricultural products, milk, is readily contaminated by radioiodine and radiocaesium.




$^{137}$Cs and $^{131}$I specific activities in milk from west central Scotland, May 1986




Average $^{137}$Cs specific activity in UK milk, 1959 – 1977

# Behaviour of Cs in the soil

- **Mineral soils** - soils containing a large proportion of layered clays retain most radiocaesium on the solid, leaving only a small amount in the soil solution thus reducing the uptake in plants.

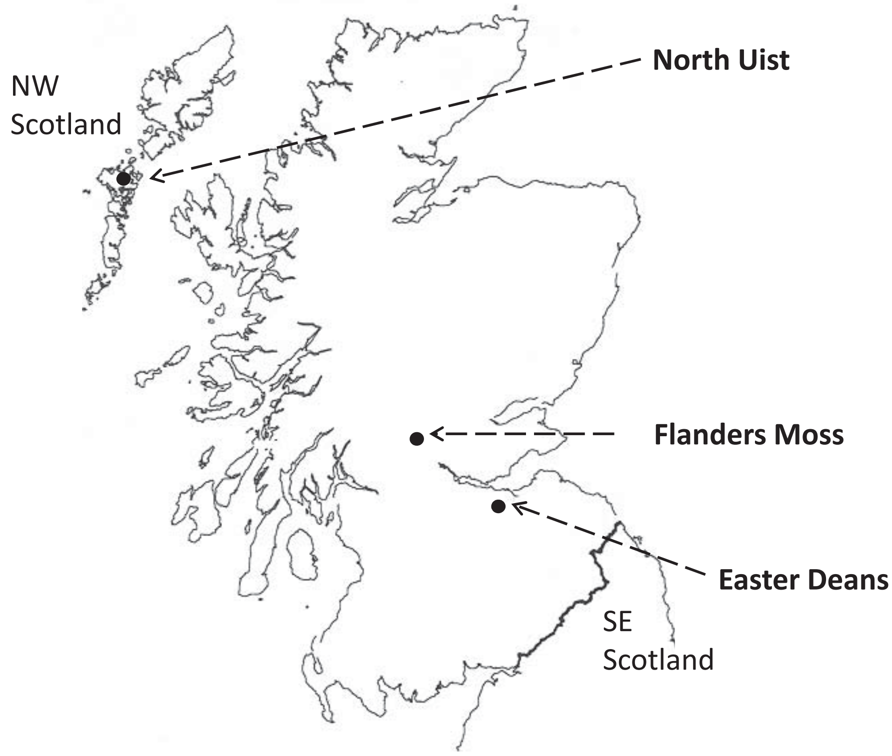
immobilisation

- **Organic soils** - soils lacking these clays allow high levels of radiocaesium in the soil solution readily available for plant uptake.

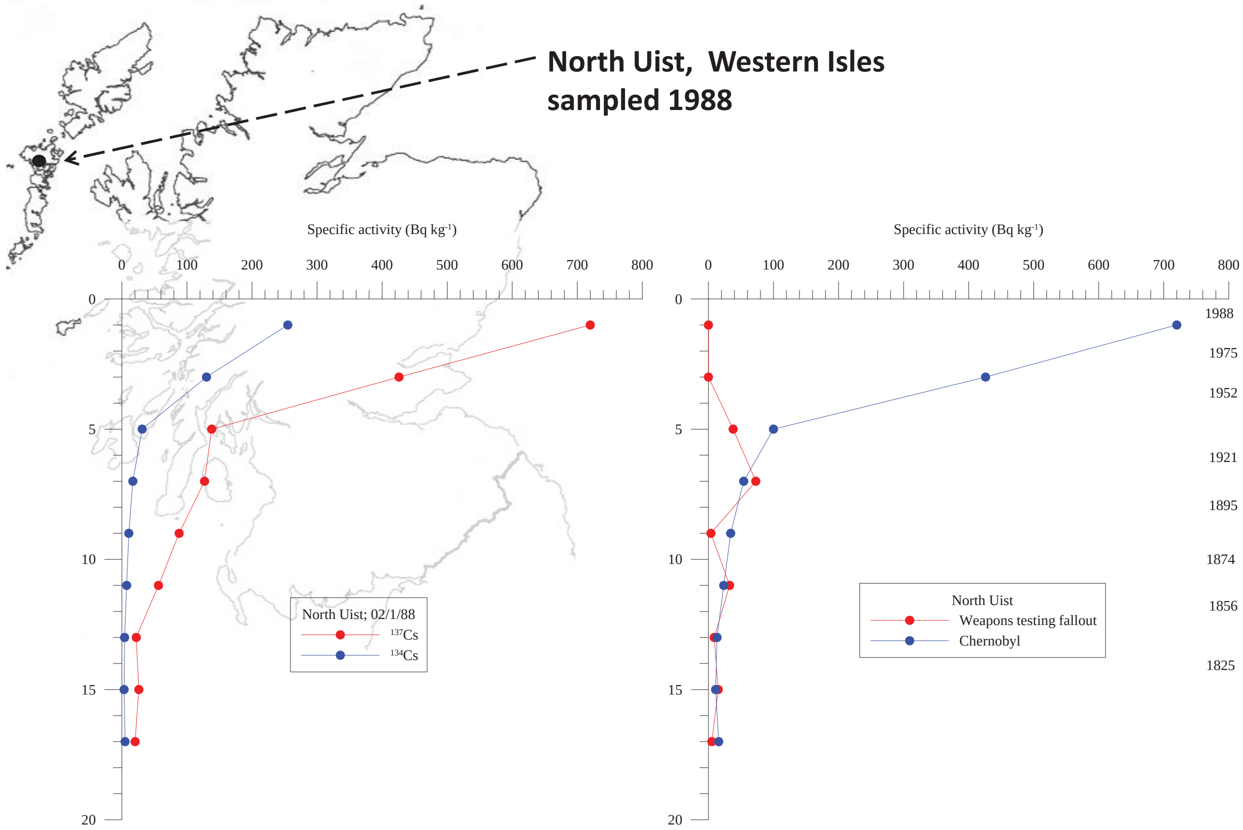
high but reversible sorption

high bioavailabilty

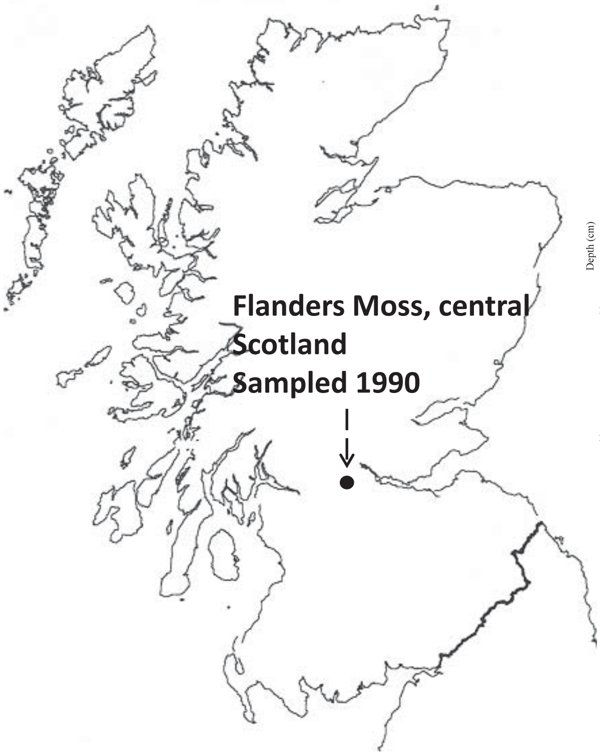
# Behaviour of Cs in the soil

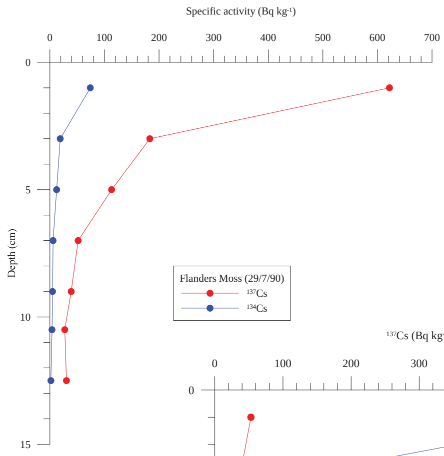


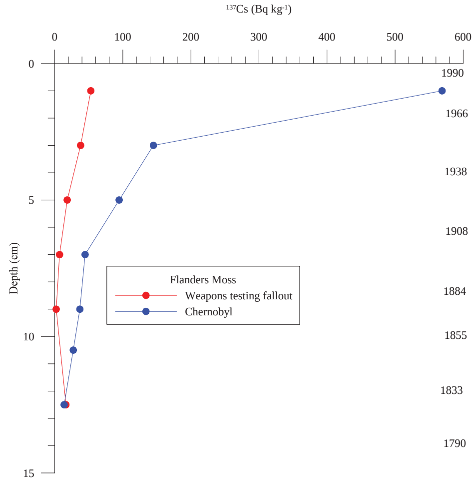

# Behaviour of Cs in the soil



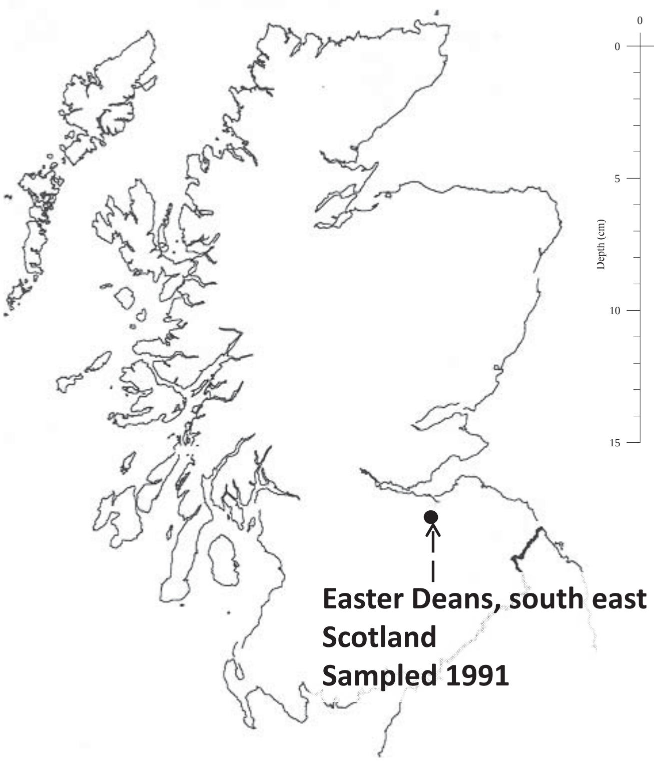
# Behaviour of Cs in the soil

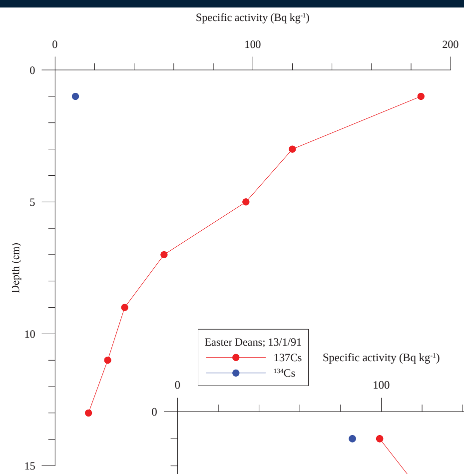


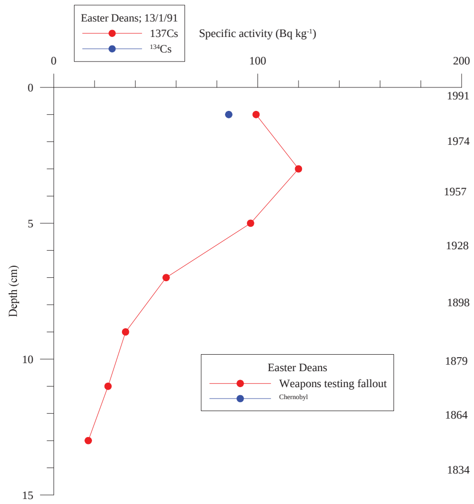




# Behaviour of Cs in the soil



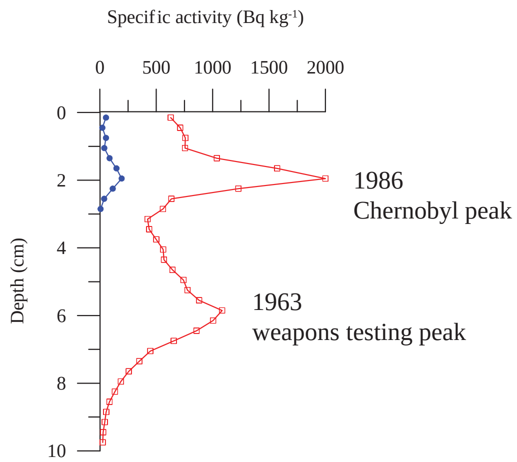



# Behaviour of Cs in the soil

| Site | Sampling date | Weapons testing inventory | Chernobyl inventory |
|---|---|---|---|
| North Uist | 02/01/1988 | 582 | 3453 |
| Flanders Moss | 15/12/1990 | 362 | 3062 |
| Easter Deans | 13/01/1991 | 1120 | 169 |

$^{137}Cs$ inventories (Bq $m^{-2}$) in Scottish peat cores

# Behaviour of Cs in freshwater




$^{137}Cs$ distribution in a sediment core from Loch Lomond
26/11/91

## Transfer of Cs from plants to animals

- In 1986, over 6770 holdings were placed under restriction for the movement and slaughter of sheep in order to prevent animals with high levels of radiocaesium entering the food chain.

- In 1995, 410 holdings were still within restricted areas and it was not until last year (in 2012) that restrictions were finally lifted.

- In the case of grazing animals, concentration rather than total deposition of Cs needs to be taken into account as the total deposition per unit area may not be high but where vegetation is sparse and the concentrations in the plant high, the animals grazing could ingest significant amounts of radionuclides.

- The concentration in individual plant species can also be important where they are the specific food of certain animals e.g. red grouse and heather.

## Cs in the Scottish Population

- DoE commissioned whole body monitoring to measure uptake of radioactivity

- The influence of sex, age, diet, and geographical location on the uptake of radiocaesium was examined in detail

- No significant difference in uptake was observed between males and females

- Children had significantly lower radiocaesium levels than adults

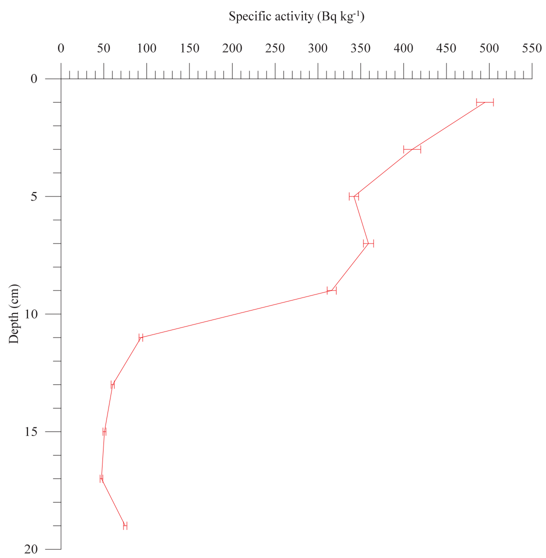
- Fresh milk consumption appeared to be the most important dietary factor influencing body levels

- Radiocaesium levels were consistently higher in the meat eating volunteers compared to vegetarians

# Cs in the Scottish Population

- Higher in west and south west and in the north and north east of the country where fallout was known to be higher
- Venison (deer meat) and goat meat eaters formed a special group with significantly higher levels than the rest of the population
- The overall picture which emerges from the study is that all members of the Scottish population took up radiocaesium to a greater or lesser extent
- Total mean dose equivalents for radiocaesium were 31 µSv for the first year after Chernobyl, 20 µSv for the second year and 62 µSv for the overall dose commitment
- In comparison with the dose due to natural background sources of radiation, the doses from radiocaesium received after Chernobyl represent increases of 1-2%

# Radionuclide legacy




Cs specific activity (Bq kg$^{-1}$) at Flanders Moss, central Scotland 2009

UK : complex atmospheric fallout & discharge sources

Fukushima : simpler system

- nuclear weapons and testing (1945 – peak in 1963)
  $^{137}Cs$
- Fukushima Dai-ichi (2011)
  diagnostic $^{134}Cs/^{137}Cs$ ratio

*…can be used as tracers to better understand the behaviour and environmental processes in complex ecosystems…*

Loch Etive, Argyll

Flanders Moss, near Stirling

Southwick saltmarsh, Solway Coast

University of Glasgow

SUERC
Scottish Universities Environmental Research Centre