30

ENVIRONMENTAL DISTRIBUTION AND TRANSPORT OF RADIONUCLIDES IN WEST CUMBRIA FOLLOWING THE WINDSCALE AND CHERNOBYL ACCIDENTS

P.J. COUGHTREY, J.A. KIRTON, N.G. MITCHELL
Associated Nuclear Services Ltd., 60 East Street, Epsom,
Surrey, KT19 8EB, UK

Tél : (44) 372 740531
telex 928 400
Fax (44) 372 741887

autre adresse COUGHTREY P.J
ANS
Eastleigh House
60 East Street, Epson
GB - Surrey KT17 1HA

ABSTRACT

Data are summarised for the pattern of deposition of Cs-137 and I-131 in West Cumbria, UK, following the Windscale and Chernobyl accidents and on the apparent environmental behaviour in terms of retention on plants, transfer from soil to plant, and transfer to milk and meat. For the 1957 accident, peak deposits of Cs-137 several kilometres south of Windscale are estimated to be in the range ~16000-40000 Bq/m^2. Peak recorded concentration of Cs-137 in milk in October 1957 was 592 Bq/l, two weeks after the accident. The ratio of concentration in milk (Bq/l) to content in herbage (Bq/m^2) was ~0.4 to 0.6 m^2/l for Cs-137 and ~0.06 to 0.1 m^2/l for I-131. Peak recorded concentration of Cs-137 in milk in May 1986 was 356 Bq/l and the corresponding ratios of concentration in milk to content in herbage were 0.37 m^2/l for Cs-137 and 0.31 m^2/l for I-131. The reason for the higher value for I-131 recorded after Chernobyl compared to that for Windscale is not clear. Whereas the initial decline in Cs-137 and I-131 content in herbage after the Windscale accident occurred with a biological half-life of ~16 d, initial loss after the Chernobyl accident was much more rapid, reflecting both the effects of rainfall and the different times of year of the two inputs. No data appear to exist for radiocaesium concentrations in meat after the Windscale accident. After Chernobyl, radiocaesium concentration ratios for soil-to-plant transfer in upland ecosystems ranged from 1 to 10. Radiocaesium concentrations in sheep at some locations did not show any substantial decline from 1986 to 1989. Experience on soil-plant-animal transfer after the Chernobyl accident, coupled with measurements on contents of radiocaesium in soil from the 1950s and 1970s, indicates that Cs-137 concentrations in individual lambs from a few locations in 1958 and the middle 1960s may have been similar to concentrations measured in individual animals in summer 1986.

INTRODUCTION

West Cumbria (Fig. 1) has received deposits of radioactivity since the early 1950s from several sources. These include uranium oxide particles from the Windscale piles over the period 1954 to 1957 due to damaged cartridges lodged in the outlet air ducts [1], releases of volatile fission products during the Windscale accident of 1957 [2], weapons-testing fallout [3], routine discharges to atmosphere from fuel reprocessing by British Nuclear Fuels plc. at Sellafield [4], and deposits during May 1986 following atmospheric dispersion of material from Chernobyl [5]. This paper evaluates data that exist for Cs-137 and I-131 from these sources in relation to retention by vegetation and transfer to animal products.

PRE-1957 RELEASES

The environmental impact of particles released prior to 1957 has been evaluated by Chamberlain [1] who estimated that ~12 kg of uranium was discharged containing (amongst other nuclides) ~7.4×10^{11} Bq Cs-137. Chamberlain concluded that the impact of these releases on the food chain would have been relatively less significant than for other forms of deposit because of their large particle size (i.e. 85% between 10 and 100 μm). No published information exists for Cs-137 concentrations in soils, vegetation or animal products for the 1954-1957 period. However, data for Sr-90 concentrations in soils in November 1957 were used by Booker [6] to provide estimates of Cs-137 deposits prior to 1957. These ranged from 12200 Bq/m^2 at 0.5 km to <185 Bq/m^2 at >13.1 km to the north east of Windscale.

OCTOBER 1957 RELEASES

Chamberlain [7] estimated a release of 8×10^{13} Bq Cs-137 from fuel during the October 1957 accident. Of this, ~10% was retained on the stack filter, allowing ~7×10^{13} Bq to be released to atmosphere. Assuming an overall ratio of I-131/Cs-137 of 20 (i.e. similar to that in the fuel), Chamberlain estimated that ~1.4×10^{15} Bq I-131 would have been released to atmosphere. Environmental measurements following the Windscale accident have been reported and evaluated by several authors [1,2,6,8-11]. Booker [6] used data on I-131 levels in herbage during the period 13-28 October

1957 to construct contours for the relative distribution of Cs-137 assuming an I-131/Cs-137 ratio of 50:1 (Fig. 2). His analysis showed total Cs-137 deposits reaching 7000-12000 Bq/m^2 at some locations several km to the south and south east of Windscale. Booker neglected to include the original I-131 contour to the south which equated to 16300 Bq/m^2 Cs-137 - this has been included in Fig. 2. Though Booker assumed an I-131/Cs-137 ratio of 50:1, Chamberlain (above) suggested that a ratio of 20:1 can be supported by the available data. This ratio would increase the estimated Cs-137 deposits given in Fig. 2 by a factor of 2.5, i.e. for Black Coombe from ~16000 to ~40000 Bq/m^2.

Contents of Cs-137 reported for vegetation after the Windscale accident ranged from 500 Bq/m^2 at Millom to 1440 Bq/m^2 at Seascale on 28 October 1957, and represented 16 to 37% of the total observed deposit to 4 inches depth in soil [8]. Concentrations in spot samples of herbage ranged from <100 to 36000 Bq/kg dw. For herbage at Seascale, the peak concentration (66300 Bq/kg dw) was recorded on 13 October 1957 and the effective half-life for decline in I-131 concentration over the period October to November 1957 based on deposit per unit area was ~4.9 d [8], implying a biological half-life of ~16 d. Cs-137 showed a similar pattern to that for I-131 during October and November 1957 but a relatively slow decline from January to March 1958.

Booker [8] reported a peak I-131 concentration in milk at Sellafield of 48800 Bq/l on the afternoon of 12 October 1957; subsequently concentrations declined with an effective half-life of ~4.7 d (i.e. similar to that for herbage at Seascale). At Seascale, the ratio of I-131 concentration in milk (Bq/l) to content in herbage (Bq/m^2) on 13 October 1957 was 0.06 m^2/l. Chamberlain and Dunster [9] reported a corresponding ratio of 0.1 m^2/l.

No data are available for Cs-137 concentrations in milk prior to 28 October 1957, reflecting difficulties in detecting Cs-137 in the presence of I-131 with the equipment then in use [2,12]. Samples from Seascale for 28 October 1957 showed a Cs-137 concentration of 592 Bq/l. The ratio of concentration in milk to content in herbage was 0.41 m^2/l. In a more detailed survey of milk concentrations over the period 8-11 November 1957, the maximum Cs-137 concentration recorded was 555 Bq/l at Corney (on rising ground ~15 km south of Windscale). The herbage content at this site on 28 October 1957 was ~1300 Bq/m^2. Allowing for a decline due to weathering from 28 October to 8-11 November, this implies a ratio of milk concentration to herbage content of ~0.6 m^2/l. Milk concentrations were lower than might have been anticipated because the accident occurred at a time of year when dairy cattle were receiving an increasing intake of conserved fodder harvested prior to the accident.

No published data appear to exist for I-131 or Cs-137 concentrations in meat of animals. The impact to the consumer would have been reduced because beef cattle and sheep can be expected to have been slaughtered at relatively long times after the original deposit.

WEAPONS-TESTING FALLOUT

Several publications provide information on the cumulative input of Cs-137 to soils of the UK as a result of weapons-testing fallout [3,13-15]. Fallout contributions of Cs-137 can be estimated from relationships between measured deposits and average annual rainfall. Maximum cumulative inputs to soils of West Cumbria from weapons-testing to the end of 1980 calculated on this basis are unlikely to have exceeded 6000 Bq/m^2 [11]. Cs-137 contents in soils in Cumbria in 1978 are given in Fig. 3 and show a complex pattern which combines the local influence of Sellafield, altitude, and pre- and post-1957 releases (Fig. 2).

Surveys in the UK in 1963, the period of maximum weapons-testing fallout, showed highest concentrations of Cs-137 in vegetation and animal products from unimproved upland pastures [6]. At one site in Cumbria, the reported Cs-137 concentration in vegetation in 1963 was 629 Bq/kg dw. Cs-137 concentrations in mutton from upland areas of the UK other than from Cumbria ranged from 50-200 Bq/kg during 1963. However, no published data appear to exist for Cs-137 concentrations in animals grazing unimproved pastures in Cumbria during the period of maximum fallout from weapons-testing. Studies in other parts of the UK and elsewhere generally suggested that Cs-137 concentrations in food products were dominated by recent deposits rather than by the cumulative inventory, but these studies were for soils with an appreciable mineral content.

ROUTINE RELEASES FROM SELLAFIELD

Maximum annual Cs-137 discharges to atmosphere from Sellafield (4.5×10^{9} Bq) occurred in 1980 [4] and corresponded with a maximum recorded concentration in milk of 13 Bq/l for farms within 3 km of the site. Routine discharges are small when compared with the 7.4×10^{11} and 7×10^{13} Bq released prior to 1957 and during the 1957 accident (above). By April 1986, concentrations in milk had declined to only a few Bq/l at most.

DEPOSITION FROM CHERNOBYL

Results of monitoring in the vicinity of Sellafield during May 1986 have been discussed by Jackson et al [17]. There is evidence to suggest that a large fraction of I-131 and Cs-137 initially intercepted by vegetation was removed rapidly, possibly as a result of rainfall. Data for a farm at Beckermet (within 2 km of Sellafield) collected over the period 3-12 May 1986 showed a total Cs-137 inventory in soil of 15000 Bq/m^2 [18]. For four farms within 1.5 km of the works sampled over the period 1979-1982, Bradford et al [19] reported Cs-137 contents in root matt plus soil in the range 9900 to 53000 Bq/m^2. Clearly, a significant fraction of the 15000 Bq/m^2 measured in 1986 must have been attributable to sources other than Chernobyl. A similar conclusion emerges for sites at greater distance to the south of Sellafield. For example, a location near Bootle showed ~12000 Bq/m^2 on 13 May 1986. Nearby locations showed measured contents of 6600 Bq/m^2 in 1957, 7800 Bq/m^2 in 1961 and 9700 Bq/m^2 in 1977.

Though it was recognised during 1986 that Cs-137 contents in soils of upland areas of West Cumbria were in the range 20000 to 30000 Bq/m^2, it was not until summer 1988 that the full extent of the contamination became clear as a result of an aerial survey [20]. This showed a central spine of Cs-137 contamination at >30000 Bq/m^2 (Fig. 4a). There were, however, some inconsistencies between the patterns observed for Cs-137 and Cs-134, particularly in upland areas to the south of Sellafield, where the aerial survey indicated Cs-137 inventories in the range 30000-50000 Bq/m^2 in some locations but corresponding Cs-134 inventories in the range 9000-12000 Bq/m^2 (Fig. 4b). Such inconsistencies were not entirely unexpected, especially if much of the Cs-137 present in the 1950s (Fig. 2) and in the 1970s (Fig. 3) remained in situ and was not removed by leaching or erosion. Naturally, the aerial survey results do not provide any indication of the availability of radiocaesium.

The maximum I-131 concentration in milk recorded at farms near Sellafield during May 1986 was ~370 Bq/l [21]. This corresponded to an initial content on pasture of 1200 Bq/m^2 giving a milk concentration to pasture content ratio of 0.31 m^2/l. The maximum Cs-137 concentration in milk of 356 Bq/l occurred on 5 May 1986 and corresponded to a pasture content of 970 Bq/m^2 on 3 May 1986, giving a milk concentration to pasture content ratio of 0.37 m^2/l. Since the Chernobyl deposit occurred at a time of year at which animals were just beginning to graze fresh pastures, the patterns of concentration in milk at different farms were highly variable according to agricultural practice [21].

DISCUSSION

Concentrations of Cs-137 and I-131 recorded in milk in October/November 1957 after the Windscale accident were higher than for May 1986 after the Chernobyl accident. For I-131, this reflects a relatively low I-131/Cs-137 ratio for the Chernobyl deposit in Cumbria. For Cs-137, the explanation appears to be the relatively small fraction retained by herbage after the Chernobyl deposit [17,18]. The ratio of concentration of Cs-137 in milk to content on herbage observed after the Chernobyl accident (0.37 m^2/l) is close to that observed after the Windscale accident (0.41 m^2/l, but note that measurements of Cs-137 were not available until some time after the 1957 accident). Similar ratios after Chernobyl and Windscale contrast with the markedly lower equilibrium transfer factors (f_m) derived post-Chernobyl relative to weapons-testing fallout and tracer experiments [5]. For I-131, the ratio observed after Chernobyl (0.31 m^2/l) was considerably higher than that observed after Windscale (0.06 to 0.1 m^2/l). The reasons for this are not clear but could involve either differences in the relative contributions of organic and inorganic forms in the two source terms, or differences in dietary intakes between spring and autumn.

By far the major impact of the Chernobyl deposit in West Cumbria has been the effect on sheep which graze upland areas. Cs-137 concentrations in sheep in some areas did not decline from 1986 to 1989. This observation is particularly important since it implies that the cumulative inventory of Cs-137 in soil is an important aspect of the environmental consequences of radiocaesium contamination in UK uplands. Given the fact that some upland areas of West Cumbria contained significant inventories of Cs-137 prior to the Chernobyl accident, it is interesting to speculate on the relative significance of the various components of these inventories.

The direct method for assessing the significance of the various components requires detailed information on the past history of Cs-137 deposits and assumptions concerning the extent to which radiocaesium remains available in soil or is removed from the rooting zone via erosion or leaching. There is emerging evidence from studies in the UK and Scandinavia that, in highly organic soils, radiocaesium remains available for plant uptake but not for physical or chemical removal processes [5]. Fells to the south of Windscale received maximum deposits of Cs-137 in 1957 in the range 16000 to 40000 Bq/m^2. Some of these fells are dominated by *Calluna vulgaris* (heather) which is known to be particularly effective at recycling radiocaesium and retaining it in surface layers [22,23]. Making the optimistic assumption that losses from soil represented 50% of the original input and taking into account radioactive decay, the maximum 1957 deposit would have reduced to 4000-10000 Bq/m^2 by 1986. Maximum weapons-testing fallout accumulated by 1986 and corrected for radioactive decay is unlikely to have exceeded 6000 Bq/m^2 (equivalent to 2 m rainfall), giving a maximum total inventory by 1986 of 10000-16000 Bq/m^2.

The second method makes use of the input of Cs-134 with the Chernobyl deposit and requires assumptions concerning the Cs-137/Cs-134 ratio in the initial deposit for the area considered. On a UK basis, this ratio appears to have been rather variable (a reasonable range being 1.5 to 2.0 [5]). However, when data for Cumbria alone are examined, the reported values include [11]: 1.59 (range 1.51 to 1.65) for air in the vicinity of Sellafield; 1.53 for rainwater at Sellafield; and 1.5 for water from two lakes. To these data can be added the value of 1.55 (range 1.35 to 1.79) for rainwater from South Cumbria [ITE, unpublished]. Assuming a ratio of 1.6 for the initial deposit in Southwest Cumbria, no more than 50% of the Cs-137 inventories measured in upland soils during 1986 at the most contaminated sites can be attributed to Chernobyl [23]. A maximum Cs-137 content in soil of 34500 Bq/m^2 was observed under *Calluna vulgaris* on fells to the south of Sellafield during November 1987 [23]. Correcting for radioactive decay, and assuming an initial Cs-137/Cs-134 ratio of 1.6 (range 1.8 to 1.4), indicates a pre-Chernobyl Cs-137 content at this site of 16800 (range 14600 to 19100) Bq/m^2. This corresponds with the maximum value estimated above by the direct method.

Unfortunately, if the second method is applied to vegetation samples collected from 1986 to 1989, it becomes clear that some plant species must have had an appreciable concentration of Cs-137 prior to 1986 [24]. These species include native herbs and shrubs typical of acid grasslands in the UK (i.e. *Eriophorum vaginatum*, *Trichophorum caespitosum*, *Calluna vulgaris*, *Narthecium ossifragum* and species of *Carex*) [23-25]. For these species, soil-to-plant transfer factors observed in 1986 and subsequently were typically in the range 1-10 [26,27]. As a result, Cs-137/Cs-134 ratios derived for individual plant species varied quite widely according to the relative contributions of Chernobyl and other sources to the radiocaesium inventory in soil at different depths in the rooting zone [11,23].

Both methods imply that maximum Cs-137 contents in soil at selected upland locations in West Cumbria prior to the Chernobyl accident may have been in the order of 15000 Bq/m^2. Using the conservative assumption that this inventory was dispersed in the top 15 cm of soil, assuming a soil bulk density of 0.5 g/cm^3 and a soil-to-plant transfer factor in the range 1-10, the estimated Cs-137 concentration in vegetation would have been 200-2000 Bq/kg dw. Assuming a transfer coefficient for lamb of 0.6 d/kg and a daily dry matter intake of 0.6 kg/d, the estimated range of maximum concentrations in individual lambs would have been 70-700 Bq/kg fw. On the basis of the observed continuing availability of the Chernobyl deposit in upland areas, the pattern of Cs-137 concentration in sheep prior to the Chernobyl accident can be expected to have followed the cumulative Cs-137 content in soil.

Taking into account the higher concentrations of Cs-137 reported for milk in 1957 compared with those reported for 1986 and the known pattern of input of weapons-testing fallout, then maximum concentrations of Cs-137 in sheep after the 1957 accident and in the middle 1960s will have been similar to or higher than maxima recorded after the Chernobyl deposit. To our knowledge, this factor has not been considered when assessing the relative radiological impacts of the Windscale and Chernobyl accidents to the population of West Cumbria.

REFERENCES

1. Chamberlain, A.C. Environmental impact of particles emitted from Windscale piles. AERE-R12163. Atomic Energy Authority, Harwell, 1986.

2. Dunster, H.J., Howell, H. and Templeton, W.L. District surveys following the Windscale accident, October 1957. Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Part 19, pp. 296-308. UN, Geneva, 1958.

3. Cawse, P.A. Studies of environmental radioactivity in Cumbria, part 4. caesium-137 and plutonium in soils of Cumbria and the Isle of Man. AERE-R9851. Atomic Energy Authority, Harwell, 1980.

4. British Nuclear Fuels plc.. Annual Reports on Radioactive Discharges and Monitoring of the Environment. Health and Safety Directorate, Risley.

5. Coughtrey, P.J., Jackson, A., Beetham, C.J. and Cuff, Y.S. Radionuclide distribution and transport in terrestrial and aquatic ecosystems after the Chernobyl disaster. ANS 2370-R1. Associated Nuclear Services Ltd., Epsom, 1990.

6. Booker, D.V. Caesium-137 in soil in the Windscale area. AERE-R4020. UK Atomic Energy Authority, Harwell, 1962.

7. Chamberlain, A.C. Comparisons of the emissions in the Windscale and Chernobyl accidents. AERE-M3568. UK Atomic Energy Authority, Harwell, 1987.

8. Booker, D.V. Physical measurements of activity in samples from Windscale. AERE/HP/R2607. UK Atomic Energy Authority, Harwell, 1958.

9. Chamberlain, A.C. and Dunster, H.J. Deposition of radioactivity in north west England from the accident at Windscale. Nature (Lond.) 182:629-630, 1958.

10. Chamberlain, A.C. Deposition of iodine-131 in Northern England in October 1957. Nature (Lond.) 183:360-361, 1959.

11. Coughtrey, P.J. A review of levels and distribution of caesium in upland environments prior to the Chernobyl accident. ANS 2131-R1. Associated Nuclear Services Ltd., Epsom, 1989.

12. Chamberlain, A.C. Relation between measurements of deposited activity after the Windscale accident of October, 1957. AERE HP/R-2606. UK Atomic Energy Authority, Harwell, 1958.

13. Cambray, R.S. and Eakins, J.D. Studies of environmental radio-activity in Cumbria, part 1. Concentrations of plutonium and caesium,-137 in environmental samples from west Cumbria and a possible maritime effect. AERE-R9807. UK Atomic Energy Authority, Harwell, 1980.
14. Eakins, J.D., Pattenden, N.J., Cambray, R.S., Lally. A.E. and Playford, K. Studies of environmental radioactivity in Cumbria, part 2. Radionuclide deposits in soil in the coastal region of Cumbria. AERE-R9873. UK Atomic Energy Authority, Harwell, 1981.
15. Cawse, P.A. and Horrill, A.D. A survey of caesium-137 and plutonium in British soils in 1977. AERE-R10155. UK Atomic Energy Authority, Harwell, 1986.
16. Agricultural Research Council Radiological Laboratory. Annual Reports for 1961-1962, 1962-1963, 1963-1964 and 1964-1965. ARCRL-8, -10, -12 and -14. ARC, Letcombe, 1962-1965.
17. Jackson, D., Jones, S.R., Fulker, M.J. and Coverdale, N.G.M. Environmental monitoring in the vicinity of Sellafield following the deposition of radioactivity from the Chernobyl accident. J. Soc. Radiol. Prot. 7:75-87, 1987.
18. Fulker, M.J. Aspects of environmental monitoring by British Nuclear Fuels plc following the Chernobyl reactor accident. J. Environ. Radioactivity 5:235-244.
19. Bradford, W.R., Curtis, E.J.C. and Popplewell, D.S. Radioactivity in environmental samples taken in the Sellafield and Ravenglass areas of West Cumbria. Sci. Total Environ. 35:267-283, 1984.
20. Sanderson, D.C.W. and Scott, E.M. Aerial radiometric survey in West Cumbria 1988. Final Report Project N611. Scottish Universities Research and Reactor Centre, East Kilbride, 1988.
21. Coughtrey, P.J., Jones, C.H., Mitchell, N.G. and Smith, A.D. Radionuclide uptake in foodstuffs produced in the UK following the Chernobyl incident - model evaluations and field data. ANS 750-R1. Associated Nuclear Services Ltd., Epsom, 1986.
22. Bunzl, K. and Kracke, W. Distribution of Pb-210, Po-210, stable lead and fallout in soil, plants and moorland sheep of a heath. Sci. Total Environ. 39:143-159, 1984.
23. Coughtrey, P.J. Models for radionuclide transport in soils-implications of the Chernobyl accident. Paper presented at the All Union Conference on Landscape Geochemical Aspects of Radionuclide Migration. Suzdal, 13-17 November 1989.

24. Coughtrey, P.J., Kirton, J.A. and Mitchell, N.G. Caesium transfer and cycling in upland pastures. Sci. Total Environ. 85:149-158, 1989.
25. Coughtrey, P.J., Kirton, J.A. and Mitchell, N.G. caesium distribution and cycling in upland pastures of N. Wales and Cumbria. Sci. Total Environ. (in press).
26. Kirton, J.A., Coughtrey, P.J. and Mitchell, N.G. derivation of soil to plant concentration ratios for upland plant species in the glasshouse and comparison with field and experimental measurements. Sci. Total Environ. (in press).
27. Coughtrey, P.J., Kirton, J.A., Mitchell, N.G. and Morris, C. Transfer of radioactive caesium from soil to vegetation and comparison with potassium in upland grasslands. Environ. Pollut. 62:281-315.

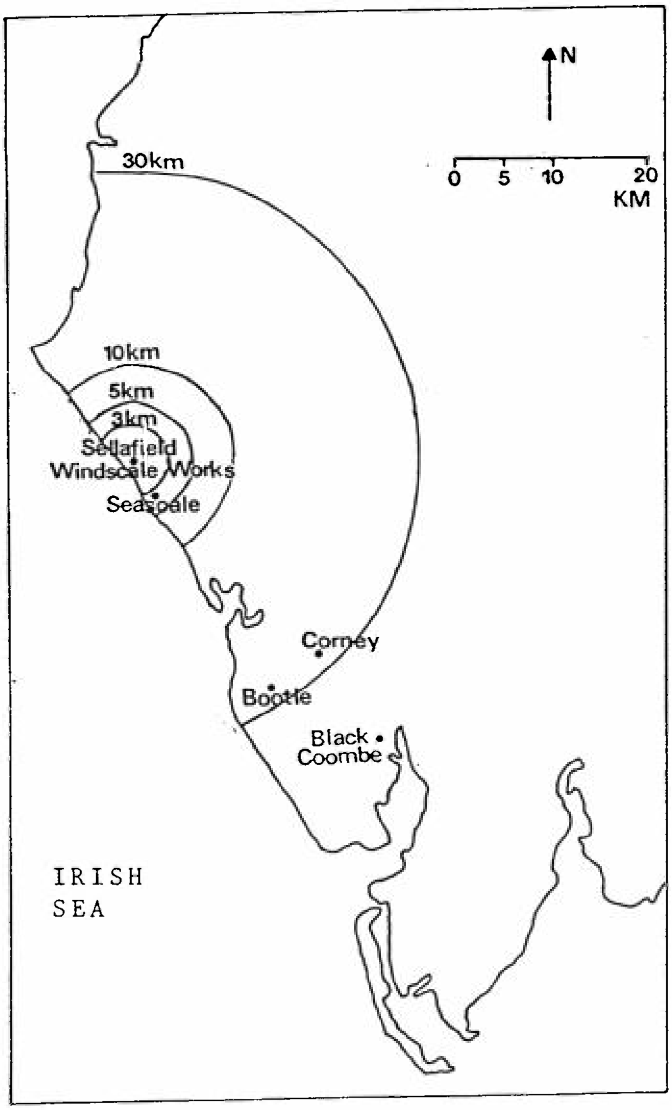
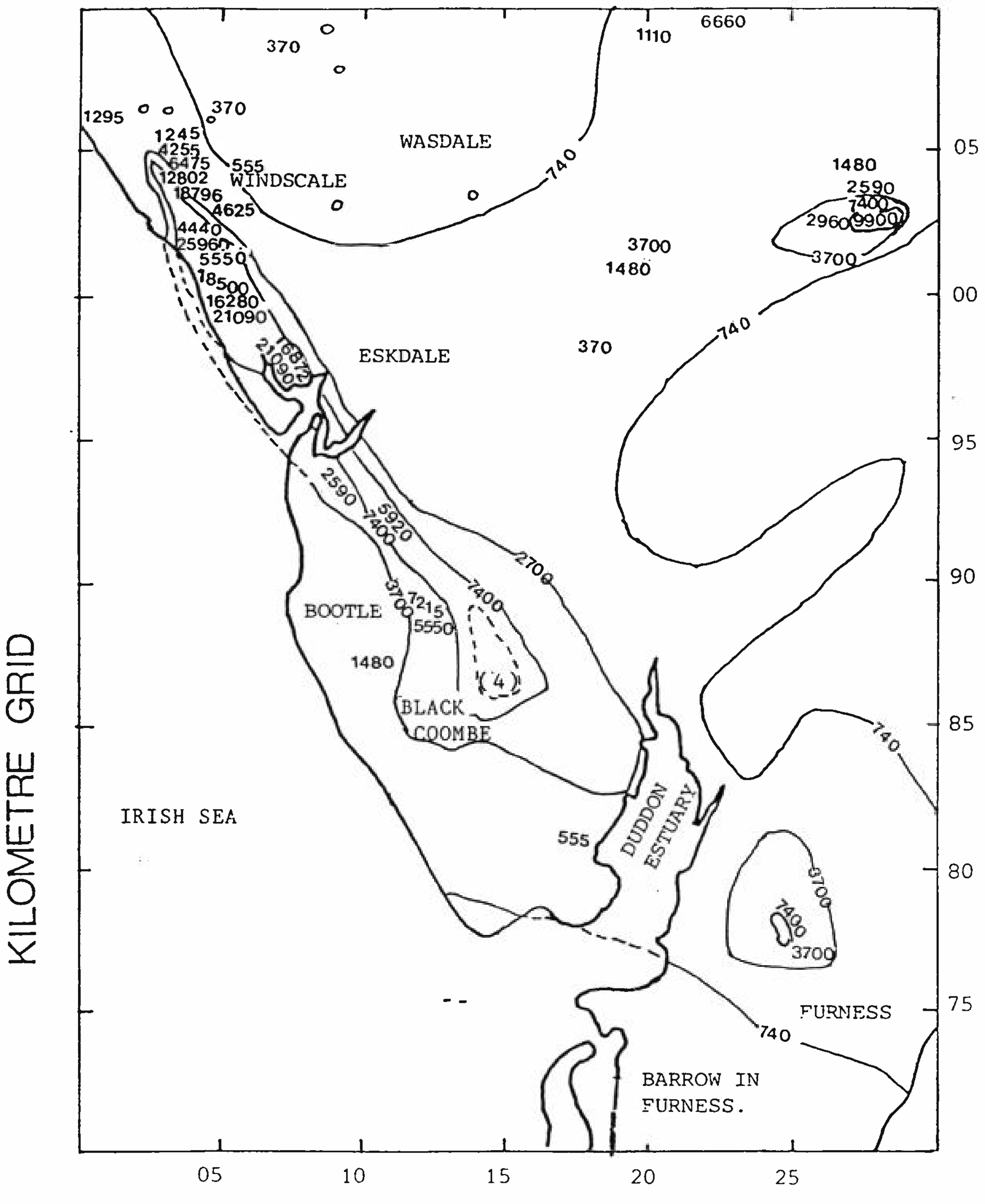


FIGURE 1. LOCATION OF SITES REFERRED TO IN TEXT.



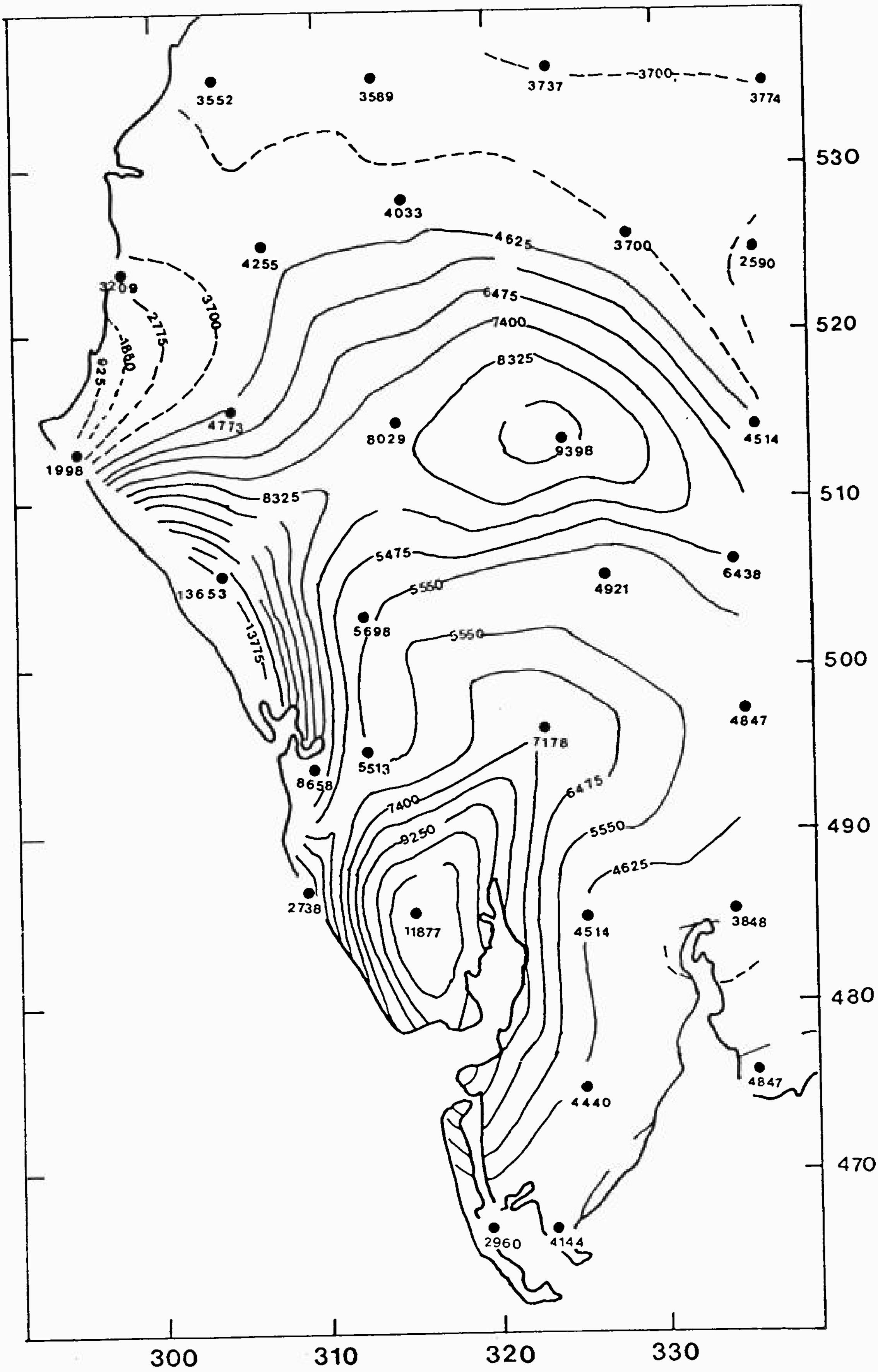
Notes 1. After Booker [6]

2. Figures are Bq m^{-2} of Cs-137 in soil

3. Contours are Bq m^{-2} reproduced from I-131 contours x 0.02

4. Additional contour based on observed I-131 deposits equivalent to 11,100 Bq m^{-2} Cs-137 assuming I-131/Cs-137 ratio = 50/1.

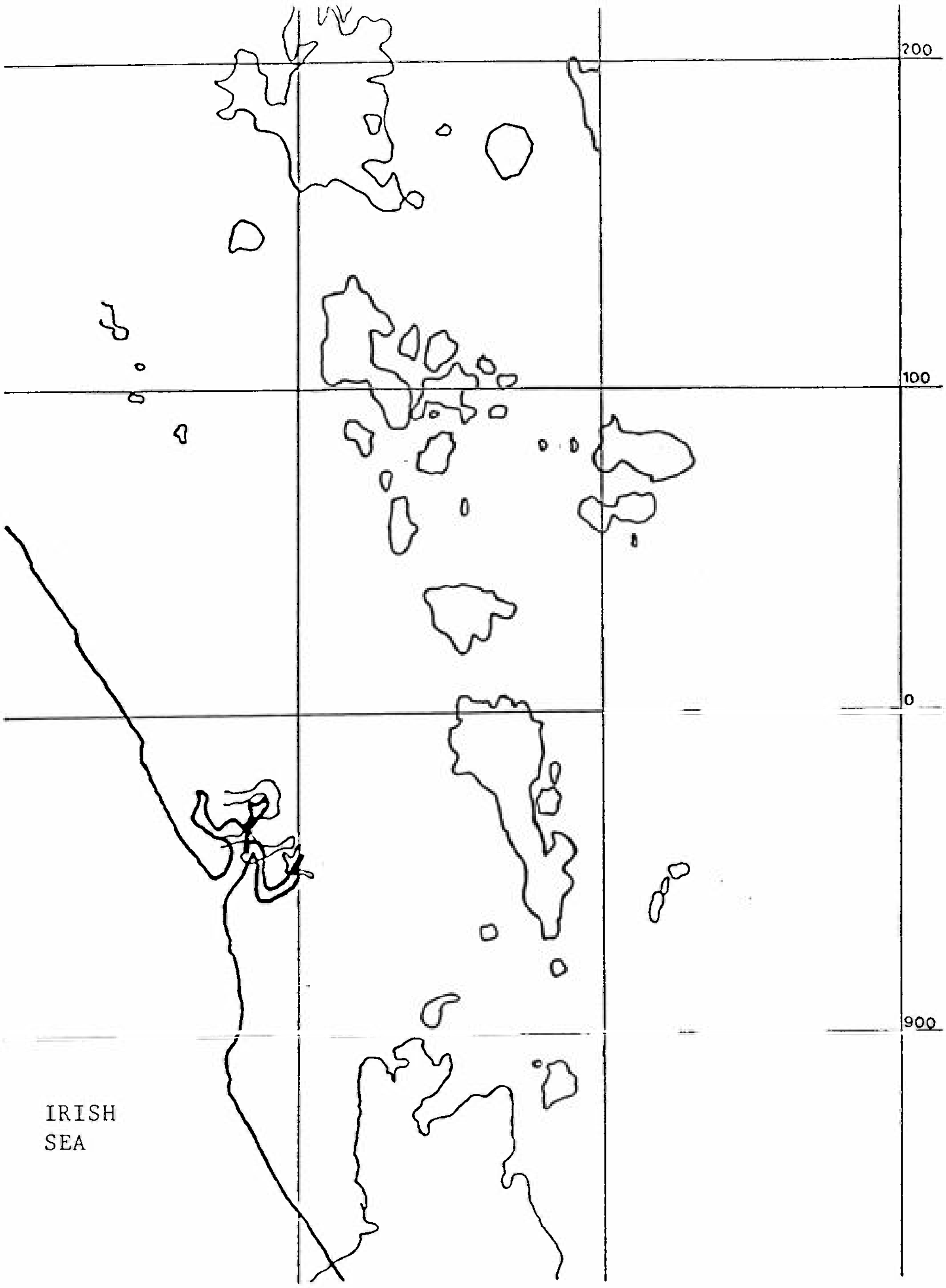
FIGURE 2. PATTERN OF DEPOSITION OF Cs-137 DUE TO THE WINDSCALE ACCIDENT.



Notes 1. From Cawse [3]

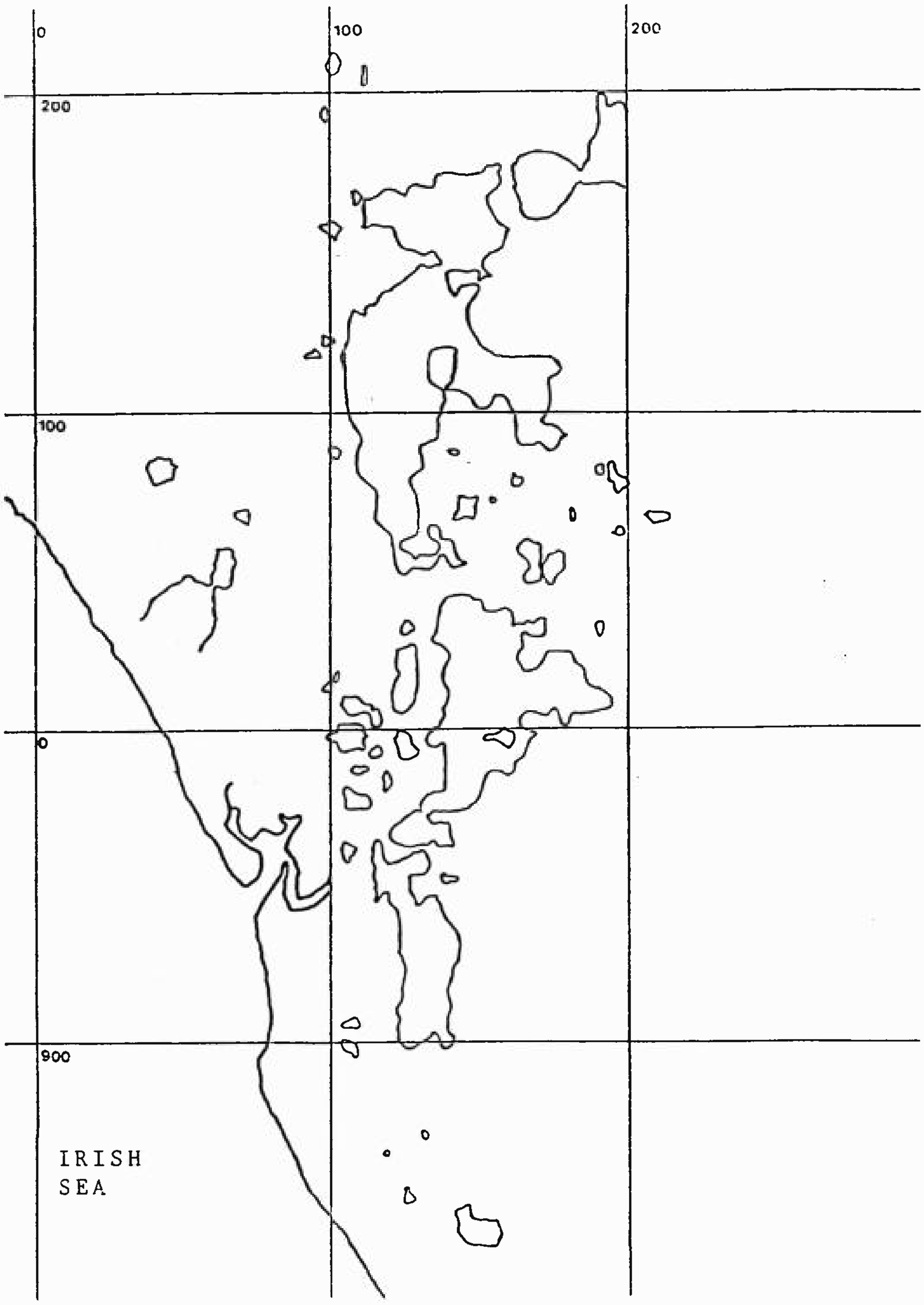
2. Deposits given as Bq m^{-2}

FIGURE 3. DEPOSITS OF Cs-137 TO 15 CM DEPTH IN THE 1978 GRASSLAND SURVEY.



Notes: 1. From Sanderson and Scott [20]
2. Contour shows Cs-137 > $30 kBq/m^2$

FIGURE 4a Cs-137 GROUND DEPOSIT IN CUMBRIA IN NOVEMBER 1988



7

Notes 1. From Sanderson and Scott [20]

2. Contour shows Cs-134 > 12kBq/m²

FIGURE 4b. Cs-134 GROUND DEPOSITS IN CUMBRIA IN NOVEMBER 1988.