Radiocaesium in the seas of northern Europe: 1980-84

W. C. Camplin and A. K. Steele

MAFF

Fisheries Research Data Report No 25
Directorate of Fisheries Research, Lowestoft, 1991

MINISTRY OF AGRICULTURE, FISHERIES AND FOOD
DIRECTORATE OF FISHERIES RESEARCH

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LOWESTOFT
1991

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Data Rep., MAFF Direct. Fish. Res., Lowestoft, (25), 174pp.

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FOREWORD

It was a shock to us all at the Fisheries Laboratory in Lowestoft when Ken Steele died on 20 August 1990.

He joined MAFF in 1958 and worked on radioactivity in the environment for more than thirty years, firstly at the Hamilton Dock Laboratory in Lowestoft, and more recently at the new extension to the main Fisheries Laboratory. His contribution to the subject is not only represented by the publications and many internal reports which he produced, but by his central role in field work and on cruises, which provided most of the samples of sea water for analysis.

At the time of his death, his knowledge of the behaviour of radiocaesium in shelf-seas of northern Europe was second to none in the Laboratory. Without his help, it would not have been possible to produce this report. It, and subsequent issues in the series, stand as a tribute to his labours.

He will be sadly missed by his fellow scientists, his family and his friends.

D. J. Garrod
Director
Ministry of Agriculture,
Fisheries and Food
Directorate of Fisheries Research

SUMMARY

A computer data base of measurements of caesium-134 and caesium-137 in sea water has been compiled. The measurements were carried out by the UK Ministry of Agriculture, Fisheries and Food and include data for the Irish Sea, Bristol Channel, English Channel, North Sea, Scottish waters and other parts of the marine environment of northern Europe. This report covers the sampling period 1980-84 and further information will be published as the data base is expanded. It is hoped that the information will be useful to scientists modelling radionuclide dispersion in the area. In order to assist in the development of new models, diskettes of the data in ASCII files are available, on request, from the first author. The data include the locations of sample collection, collection date, depth, salinity, concentration of activity and counting errors. Plots of the geographical scope of the data are also provided to assist the user.

1. INTRODUCTION

Waste disposal operations of the nuclear industry, which must be authorised under the Radioactive Substances Act (Great Britain—Parliament, 1960), may introduce novel tracers into the sea. These can be used to study sea-water movements and, ultimately, to build or validate hydrographic models. Radiocaesium is particularly useful in this context because it largely remains in solution in sea water. Many other radionuclides can be particle reactive substances and the processes which govern their distribution can be more complex.

The use of radiocaesium in studies on dispersion is not a new procedure. Preston *et al.* (1971) presented results of measurements in sea water in the Irish Sea during the 1960s. They derived contours for the concentrations of caesium-137 in sea water close to the Sellafield site and normalised these to unit discharge rate. The normalised quantity was used to assess the maximum permissible discharge from the site; these limits were compared with those derived using a simple, pre-operational, dispersion model. Agreement was found to be good and it was concluded that detailed studies of dispersion can often be dispensed with in favour of simple models which provide an adequate basis for planning purposes.

Modelling has developed a great deal since that time. A simple model in the 1990s is one such as the box model for the Irish Sea and coastal waters of northern Europe published by Jefferies and Steele (1989). This model began its development as a series of compartments whose exchanges were estimated from flow rates derived using methods which were not connected with radioactivity. It soon developed into a semi-empirical model using caesium-137 as a tracer to tune the predicted concentrations in sea water. In the paper by Jefferies and Steele, the tuning was carried out by adjusting the exchanges by trial and error. A more sophisticated method had been used by Hallstadius *et al.* (1987) on a similar box model. Transfer coefficients were calculated by a computer program to give the best fit, in a least-squares sense, between simulated and actual concentrations in sea water. Similar models have been published by, for example, Clark *et al.* (1980), Camplin *et al.* (1982), McKay and Baxter (1985) and Jones (1990).

More complex hydrodynamic models are also being developed for the coastal seas of northern Europe (Pohlmann *et al.*, 1987). Here too, radiocaesium data have been used to validate model predictions and, in this case, this has been carried out independently of the parameterisation of the processes modelled.

Several laboratories have reported the results of measurements of radiocaesium in sea water. Most, if not all, sea areas in northern Europe have been studied, (MAFF, 1989; CCRX, 1986;

Mitchell *et al.*, 1988; Aarkrog *et al.*, 1988; Bettencourt *et al.*, 1988; Nies, 1990 and Guegueniat *et al.*, 1988). Camplin and Aarkrog (1989), acting as rapporteurs on behalf of Working Group 2 of the Commission of the European Communities' project MARINA, collected together measurements made by several laboratories and produced a synthesis spanning the period 1980-85. The data were sorted into geographical regions on the basis of ICES fishing areas. Whilst this data set has advantages because it brings together measurements from several laboratories, it was not possible to present individual results. Information on variations within an ICES area was therefore lost to the reader and further positional data would be needed for model development and validation.

The purpose of this series of reports is to fully document the results of measurements of radiocaesium in sea water, carried out by the Fisheries Laboratory of the Ministry of Agriculture, Fisheries and Food (MAFF) in the United Kingdom. MAFF has operated an extensive programme of sampling sea water around the British Isles and beyond since the early 1960s. The data have not been published in full before and, because of the large size of the data base, we have decided to divide the information into several time periods. This report is the first of these and includes results for the period 1980-84 inclusive. Further reports will be published in due course.

The data set included in this report is available on request on diskette from the first author.

The report lists a brief description of the sampling methods used (Section 2) and the analytical procedures adopted (Section 3). The scope of the data set is described (Section 4) with the aid of figures showing the sampling locations.

2. METHODS OF SAMPLING AND PREPARATION

Various methods of sampling have been used to collect sea water. Surface samples were collected offshore from ships and at the shore. The offshore samples were obtained either by pumping through hoses or a dedicated shipboard supply, or by submerging water sampling bottles. Shoreline samples were simply collected using a bucket from within a few metres of the water's edge. Sub-surface samples were collected using large volume Niskin bottles which were triggered at the appropriate depth using mechanical or electro-mechanical means.

The sample size for radiocaesium analysis normally varied between 10 and 50 l depending on the anticipated concentration of radiocaesium in the sample. Sub-samples of approximately 250 ml were generally taken for salinity analysis. The radiocaesium samples were filtered through either 0.22 µm filter papers or 2 µm filter cartridges to remove particulates and then acidified to a pH of between 1 and 2 using nitric or hydrochloric acid, depending on the method of ion exchange to be used. Reducing the pH of the sample facilitates the selective absorption of caesium onto the resin during analysis. The samples were stored for periods between a few hours to a year depending on the availability of subsequent processing facilities.

3. MEASUREMENT OF ACTIVITY, AND SALINITY

The measurement of activity in the samples is described by Baker (1975) and Dutton (1970). It involves passing the acidified filtrate through cartridges of either ammonium-duodeca-

Mitchell *et al.*, 1988; Aarkrog *et al.*, 1988; Bettencourt *et al.*, 1988; Nies, 1990 and Guegueniat *et al.*, 1988). Camplin and Aarkrog (1989), acting as rapporteurs on behalf of Working Group 2 of the Commission of the European Communities' project MARINA, collected together measurements made by several laboratories and produced a synthesis spanning the period 1980-85. The data were sorted into geographical regions on the basis of ICES fishing areas. Whilst this data set has advantages because it brings together measurements from several laboratories, it was not possible to present individual results. Information on variations within an ICES area was therefore lost to the reader and further positional data would be needed for model development and validation.

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molybdophosphate on silica gel (ASG) or potassium cobaltihexacyanoferrate (KCFC). KCFC was chosen when there was the possibility of the presence of significant concentrations of zirconium-95 and niobium-95 in sea water. These radionuclides are absorbed by ASG and their radiations can interfere with those of caesium-134. The cartridges were counted for activity using a gated NaI (Tl) well crystal detector system. Counting times varied between 100 and 400 minutes depending on the predicted level of activity in the sample. The limits of detection varied but were generally in the range of 1-10 Bq m^{-3}.

Quality assurance of the detection method included regular recalibration of the detector using standard sources of known strength supplied by Amersham International plc, Amersham, UK, and participation in intercomparison exercises organised by the International Atomic Energy Agency (IAEA, 1988) and others (Steele, 1989).

Salinity measurements were performed using an inductively coupled Autolab Salinometer, Model 601 Mk III. Calibration was carried out using standard sea water supplied by the Institute of Oceanographic Sciences, Wormley, UK. The salinometer had a precision of ±0.003, but the greatest source of error was likely to be due to methods of sampling and storage.

4. DATA FORMAT AND SCOPE

The data set is provided at Annexes 1-3 as follows:

shoreline sampling	(Annex 1);
routine offshore sampling	(Annex 2);
and non-routine offshore sampling	(Annex 3).

The data comprise the categories explained below:

Site name	A 'site' is a collection of locations where samples have been obtained. It is often named after a nuclear establishment or a particular vessel used to collect offshore samples. A 'site' does not refer to a unique geographical area.
Site number	Each 'site' is given an identifying number.
Station name	A 'station' is a name given to the position where samples have been taken. Not all positions have names.
Station number	Each 'station' within a 'site' is given a unique number.
Position	The position of each station is recorded as latitude and longitude in integer degrees, minutes and seconds.
Sample depth	The depth at which samples have been taken is given in Annexes 2 and 3 in metres. Surface samples are attributed to a nominal depth of 3 m except where more accurate information is available. All of the samples in Annex 1 were collected at the shoreline.

Station depth	For offshore positions, the depths of the stations are provided in metres. A blank entry indicates that no data are available.
Collection date	This is self explanatory.
Salinity	The salinity is recorded in practical salinity units (psu) (UNESCO, 1981) with 3 digits after the decimal point. Our judgement is that it is generally accurate to two digits after the decimal point. 'NA' means that no data are available.
Concentration of activity	The concentration of activity is given for caesium-134 and caesium-137 in Bq m^{-3}. 'NA' means that no data are available. 'ND' means that a measurement was made but that no activity was detected in the sample using the method described. If a counting error of more than 60% was obtained, an 'ND' was entered. The errors quoted are counting errors of one standard deviation expressed as a percentage of the concentrations. The concentrations are recorded with two digits after the decimal point though the accuracy of the measurement, as indicated by the counting error, is variable.
Ratio	This refers to the ratio of the concentration of caesium-137 over the concentration of caesium-134 where both radionuclides have been detected.
Sample number	This is a unique laboratory sample number for the year in which the sample was collected.

The scope of the data, which covers the period 1980-84, is described in Table 1. The shoreline positions in Annexes 1 and 2 are sorted in clockwise order around the coast of Britain, beginning at the north-eastern-most tip of the mainland. Data from cruises listed in Annex 3 are firstly grouped by cruise and are secondly sorted chronologically.

The locations of sampling positions in Annexes 1 and 2 are shown in Figures 1-5. There were too many locations from cruises listed in Annex 3 to provide station plots for all of the data in this report. However, Table 2 indicates the general area covered by each cruise in Annex 3 by reference to the ICES fishing regions. The latter are shown in Figure 6. Station plots for data in Annex 3 are available at no charge on written request to the first author. The three annexes are available on diskette, each as a separate ASCII file with the same format as that shown in this report.

Acknowledgements

The initiative for much of this programme originated with Doug Jefferies (now retired), who worked for MAFF in Lowestoft for many years. Doug's infectious spirit and understanding of radioactivity in the aquatic environment were greatly admired by us all. Our debt to his efforts is great.

We would also like to thank the many sampling, analytical and computing staff who took a part in producing these data over the years.

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Table 1. Scope of the data in Annexes 1-3

Annex	Page	Site no.	Site name	Station no.	Station name or cruise	Position		National Grid Reference		
						Latitude	Longitude	Sheet	Easting	Northing
1	25	30	Hartlepool	1	Little Scar	54°39'57"N	1°11'4"W	93	526	304
1	25			4	PLZ	54°39'5"N	1°9'58"W	93	538	288
1	25	12	Sizewell	25	Lowestoft M/L Header Tank	52° 27'30"N	1°44'32"E	134	542	911
1	25			18	Lowestoft Beach (SE)	52°27'10"N	1°44'20"E	134	540	905
1	26			15	Sizewell Inlet	52°12'32"N	1°37'39"E	156	478	630
1	28			11	Aldeburgh	52°9'17"N	1°36'25"E	156	467	569
1	28			20	Felixstowe	51°58'24"N	1°22'52"E	169	322	360
1	28			21	Clacton	51°47'29"N	1°10'2"E	169	184	151
1	29	7	Bradwell	7	Waterside	51°43'52"N	0°53'1"E	168	991	76
1	30			118	Bradwell Inlet	51°44'40"N	0°53'35"E	168	997	91
1	30	8	Dungeness	10	Dungeness Inlet	50°54'32"N	0°57'58"E	189	85	164
1	31	18	Winfrith	6	Poole	50°41'20"N	1°59'50"W	195	6	875
1	31			2	Kimmeridge	50°36'39"N	2°7'53"W	195	907	791
1	32	22	Channel Islands	219	Alderney, East	49°44'18"N	2°11'30"W			
1	32			224	Jersey, St Catherines Bay	49°13'8"N	2°1'29"W			
1	33			216	Guernsey	49°27'24"N	2°31'30"W			
1	33	9	Hinkley	10	Inlet	51°13'2"N	3°7'47"W	182	212	471
1	34	11	Oldbury	116	Oldbury	51°38'54"N	2°35'2"W	162	596	946
1	35	6	Berkeley	115	Berkeley Inlet	51°41'37"N	2°29'52"W	162	656	996
1	36	34	Cardiff	4	Orchard Ledges East	51°27'55"N	3°7'59"W	171	213	747
1	36	14	Wylfa	6	Holyhead	53°18'59"N	4°37'44"W	114	249	832
1	37			1	Cemlyn Bay Shore	53°24'48"N	4°30'41"W	114	331	937
1	37			2	Cemaes Bay	53°24'53"N	4°27'5"W	114	371	937
1	37			3	Llandudno	53°19'29"N	3°47'24"W	116	808	824
1	38			4	Prestatyn	53°20'33"N	3°24'54"W	116	58	838
1	39	33	Capenhurst	1	PLZ (Hoylake)	53°24'39"N	3°9'20"W	108	232	911
1	39	1	Sellafield	134	New Brighton	53°26'33"N	3°3'9"W	108	301	945
1	39			51	Ainsdale	53°36'31"N	3°3'56"W	108	295	130
1	40			33	Rossal (Fleetwood)	53°54'0"N	3°2'55"W	102	311	454
1	40	29	Heysham	2	Half Moon Bay	54°2'19"N	2°54'37"W	96	404	607
1	41	1	Sellafield	74	Walney West Shore	54°7'5"N	3°16'17"W	96	169	699
1	41			52	Silecroft	54°13'1"N	3°20'53"W	96	121	810
1	42			23	Seascale	54°23'36"N	3°29'6"W	89	36	8
1	45			100	St Bees-W	54°29'20"N	3°36'26"W	89	959	116
1	47			39	W/A Shore	54°33'1"N	3°35'56"W	89	966	184
1	48			15	Maryport Shore	54°44'30"N	3°27'26"W	89	62	395
1	49			53	Silloth	54°53'1"N	3°22'41"W	85	116	553
1	49			54	Southerness	54°52'22"N	3°35'40"W	84	977	543
1	50			57	Ross Bay	54°46'35"N	4°5'46"W	83	652	444
1	50			4	Isle of Whithorn	54°41'59"N	4°21'32"W	83	480	362
1	51			2	Drummore	54°41'30"N	4°53'16"W	82	139	368
1	51			56	Portpatrick	54°50'23"N	5°7'5"W	82	998	539

Table 1. Continued

Annex	Page	Site no.	Site name	Station no.	Station name or cruise	Position		National Grid Reference		
						Latitude	Longitude	Sheet	Easting	Northing
1	52	10	Hunterston	6	Fish Farm 1	55°43'25"N	4°53'42"W	63	182	516
1	52			17	Fish Farm 2	55°43'22"N	4°53'59"W	63	179	515
1	52			18	Rearing Tank Inlet (Beach)	55°42'59"N	4°54'9"W	63	177	508
1	53	15	Dounreay	110	Sandside Bay	58°34'12"N	3°47'30"W	11	958	660
2	55	48	DAFS* Water	3	Fair Isle					
2	55		Samples	4	Buckie					
2	56			5	Aberdeen					
2	57			6	Arbroath					
2	57	37	File C	11	JONSIS 1					
2	58			12	JONSIS 2					
2	58	16	U.K. Survey	67	Lowestoft (Boat)					
2	58	22	Channel Islands	218	Jersey, Ecrehos					
2	58	37	File C	7	Swansea-Cork St B					
2	59			8	Swansea-Cork St C					
2	59			9	Swansea-Cork St D					
2	59			10	Swansea-Cork St E					
2	60	50	St Georges	1	Fishguard-Rosslare 1					
2	61		Channel	2	Fishguard-Rosslare 2					
2	62			3	Fishguard-Rosslare 3					
2	64	1	Sellafield	82	Dahn Buoy					
2	65			83	Midpoint Net Position					
2	65			116	PLZ Current Meter					
2	66	49	North Channel	1	Corsewall					
2	67			2	Mid Channel					
2	69			3	Hunter					
2	70	46	NEA Weather	1	Station A					
2	71		Ships	2	Station B					
2	72			3	Station C					
2	74			4	Station D					
2	75			5	Station E					
2	76	47	NEA Challenger	11	Challenger Mooring Y					
2	76		Line	14	Tiree Passage East					
2	76			13	Tiree Passage Centre					
2	76			12	Tiree Passage West					
2	76			1	Challenger St 1					
2	77			2	Challenger St 2					
2	77			3	Challenger St 3					
2	78			4	Challenger St 4					
2	79			5	Challenger St 5					
2	79			6	Challenger St 6					
2	80			7	Challenger St 7					
2	81			8	Challenger St 8					
2	81			9	Challenger St 9					
2	82			10	Challenger St 10					
2	82	48	DAFS* Water	1	Cape Wrath					
2	83		Samples	2	Pentland Firth					

* Now SOAFD (Scottish Office Agriculture and Fisheries Department)

Table 1. Continued

Annex	Page	Site no.	Site name	Station no.	Station name or cruise	Position		National Grid Reference		
						Latitude	Longitude	Sheet	Easting	Northing
3	85	38	Cirolana		4/80					
3	86				6/80					
3	86				8/80					
3	88				9/80					
3	89				10/80					
3	95	40	Corella/		4/80					
3	98		Dawn Sky		5/80					
3	104	38	Cirolana		6/81					
3	104				7/81					
3	120				8/81					
3	122	40	Corella/		4/81					
3	126		Dawn Sky		5/81					
3	126	38	Cirolana		3/82					
3	136				5/82					
3	139				7/82					
3	141				9/82					
3	141	39	Clione		7/82					
3	143	40	Corella/		7/82					
3	143		Dawn Sky		15/82					
3	148	38	Cirolana		5/83					
3	150				7/83					
3	150				8/83					
3	152				9/83					
3	162	39	Clione		6/83					
3	162				9/83					
3	164				11/83					
3	165	38	Cirolana		6/84					
3	165				7/84					
3	167				10/84					
3	168	39	Clione		11/84					
3	168				13/84					
3	174	51	Challenger		2/84					
3	174				10/84					

Notes:
The 'site' numbers and names are used at DFR as a convenient tool to group samples together. They do not refer to a unique geographical area. Station names are prescribed when convenient. A station is given a unique number within a 'site' group. The station numbers are not shown for cruises in this table.

Table 2. ICES areas covered by non-routine offshore sampling

Vessel	Cruise no.	Page no.	Barents Sea	Norwegian Sea	Spitzbergen Bear Island	Kattegat and Skagerrak	The Sound and Belt Sea	Baltic	North	North Sea	
										Central	South
			I	IIa	IIb	IIIa	IIIb,c	IIId	IVa	IVb	IVc
Cirolana	4/80	85									X
Cirolana	6/80	86									
Cirolana	8/80	86							X	X	X
Cirolana	9/80	88									X
Cirolana	10/80	89		X					X	X	
Corella/ Dawn Sky	4/80	95							X	X	X
Corella/ Dawn Sky	5/80	98							X	X	X
Cirolana	6/81	104									
Cirolana	7/81	104	X	X	X				X	X	
Cirolana	8/81	120							X	X	X
Corella/ Dawn Sky	4/81	122							X	X	X
Corella/ Dawn Sky	5/81	126									
Cirolana	3/82	126				X	X	X	X	X	X
Cirolana	5/82	136							X	X	X
Cirolana	7/82	139							X	X	X
Cirolana	9/82	141									
Clione	7/82	141									
Corella/ Dawn Sky	7/82	143									
Corella/ Dawn Sky	15/82	143									X
Cirolana	5/83	148							X	X	X
Cirolana	7/83	150									
Cirolana	8/83	150							X	X	X
Cirolana	9/83	152				X			X	X	X
Clione	6/83	162								X	
Clione	9/83	162									
Clione	11/83	164									
Cirolana	6/84	165									
Cirolana	7/84	165									
Cirolana	10/84	167							X	X	X
Clione	11/84	168									
Clione	13/84	168							X	X	X
Challenger	2/84	174									
Challenger	10/84	174									

Table 2. Continued

Vessel	Cruise no.	Page no.	Faroe Plateau	Faroe Bank	NW Coast Scotland N Ireland	Irish Sea	W Coast Ireland Porcupine Bank	English Channel East	English Channel West	Bristol Channel	S Coast Ireland	Bay of Biscay
			Vb1	Vb2	VIa	VIIa	VIIb,c	VIId	VIIe	VIIf	VIIg-k	VIII
Cirolana	4/80	85			X		X	X			X	
Cirolana	6/80	86							X		X	X
Cirolana	8/80	86										
Cirolana	9/80	88						X	X		X	X
Cirolana	10/80	89			X	X						
Corella/ Dawn Sky	4/80	95			X	X		X	X	X	X	
Corella/ Dawn Sky	5/80	98			X	X		X	X	X	X	
Cirolana	6/81	104					X	X	X		X	
Cirolana	7/81	104										
Cirolana	8/81	120										
Corella/ Dawn Sky	4/81	122			X	X		X	X	X	X	
Corella/ Dawn Sky	5/81	126						X	X			
Cirolana	3/82	126										
Cirolana	5/82	136			X	X		X	X		X	
Cirolana	7/82	139										
Cirolana	9/82	141				X						
Clione	7/82	141				X						
Corella/ Dawn Sky	7/82	143						X	X			
Corella/ Dawn Sky	15/82	143				X		X	X	X	X	
Cirolana	5/83	148			X	X		X	X			X
Cirolana	7/83	150							X		X	
Cirolana	8/83	150										
Cirolana	9/83	152	X	X	X	X		X	X	X	X	
Clione	6/83	162										
Clione	9/83	162						X	X	X	X	
Clione	11/83	164				X						
Cirolana	6/84	165					X	X	X		X	
Cirolana	7/84	165										
Cirolana	10/84	167							X		X	X
Clione	11/84	168				X						
Clione	13/84	168			X	X		X	X	X	X	
Challenger	2/84	174				X						
Challenger	10/84	174			X							

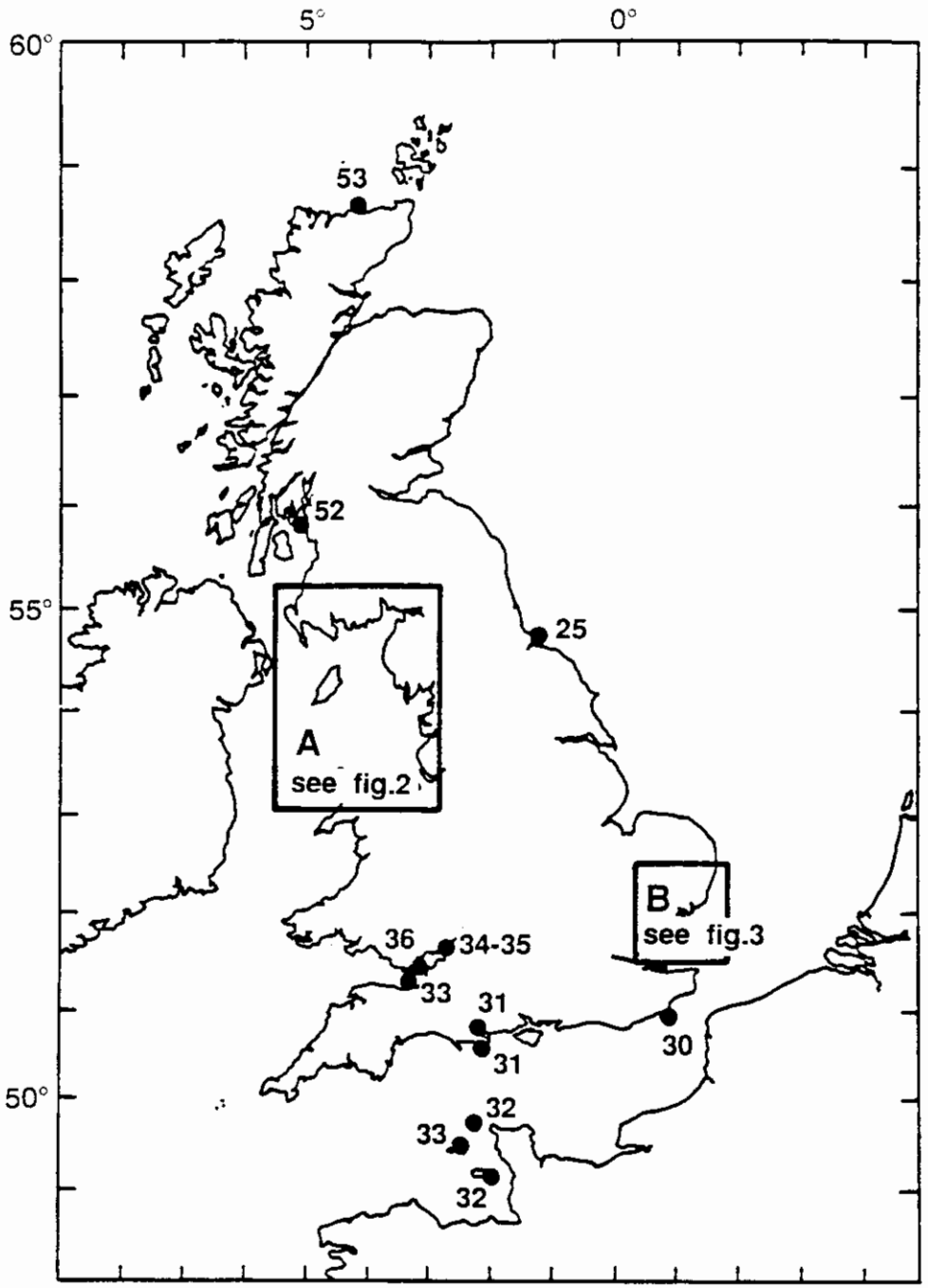

Figure 1. Location of shoreline sampling positions from Annex 1. The numbers shown on this figure refer to page numbers in the report.