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# AN EXAMINATION OF THE DILUTION OF WATERBORNE POLLUTION FROM THE GERMAN BIGHT TO THE CATTEGAT BY MEANS OF RADIOACTIVE TRACERS

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Abstract. Discharges of waterborne radioactive pollution ($^{90}Sr$, $^{134}Cs$, and $^{137}Cs$) from European nuclear reprocessing plants may be used as oceanographic tracers.

The present report shows how discharges from Cap de la Hague in France were applied to estimate the transfer of pollution from the German Bight to the inner Danish waters (The Cattegat). About 40 percent of the bottom water in the Cattegat may arise from the German Bight.

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## INTRODUCTION

The German Bight receives an important part of the waterborne pollution carried by the main north-west European rivers. The Jutland Current, which runs northward along the west coast of Jutland (cf. Fig. 1) transfers some of this pollution from the German Bight to the Cattegat. It is difficult to obtain a quantitative measure of this transport, however. One method could be to use radioactive tracers. But the radionuclides should have a relatively long half-life and be present in measurable concentrations even after a significant dilution. In the North Sea area there are two possible sources for such radionuclides: Sellafield (earlier Windscale) in the U.K. and Cap de la Hague in France. These are both reprocessing plants for spent nuclear fuel. They discharge a variety of radionuclides to the sea among these: $^{90}Sr$ with a half-life of 29 years, $^{134}Cs$ (2 years), and $^{137}Cs$ (30 years).

The present report describes how these radionuclides might be applied in order to elucidate the problem.

## METHODS AND ASSUMPTIONS

Seallafield (S) and Cap de la Hague (H) discharge their liquid waste to the Irish Sea and English Channel, respectively (Fig. 1). From the Irish Sea the radioactive effluent from (S) runs north of Scotland and then southward into the central part of the North Sea, where it is mixed with the North Sea water. Later it moves northward with the Norwegian coastal Current. A minor part enters the Danish Straits through the Skagerak. The effluent from (H) runs northward along the continental side of the English Channel, and after having passed the German Bight, it is carried further northward by the Jutland Current. Some of this pollution enters the Danish Straits,

while the remaining part is carried northward by the Norwegian coastal Current.

The discharges from (S)[1] and (H)[2] are shown in Table 1 for the periods 1976-1985 and 1978-1985, respectively. In the following calculations, all data have been decay corrected to the same date (July 1, 1983). By this, the importance of radioactive decay has been eliminated from the calculations. In other words all three radionuclides ($^{90}Sr$, $^{134}Cs$ and $^{137}Cs$) behave as stable, non-radioactive isotopes of strontium and caesium respectively.

The transfer of a substance from a source to a sample collected at a given location is expressed by the so-called transfer factor (TF). This factor is defined as the ratio of the infinite time integral of the radionuclide concentration in the sample to the total discharge of this radionuclide from the source. In other words, if a source has discharged A Bq and the time integral in the seawater at the location is B Bq $m^{-3}$ yr, the transfer factor becomes (B/A) $m^{-3}$ yr. In the steady-state case, where we assume annual discharges of $\alpha$ Bq $yr^{-1}$ and an equilibrium concentration at the location of $\beta$ Bq $m^{-3}$, the transfer factor becomes ($\beta/\alpha$) $m^{-3}yr$.

The discharges from (S) and (H) vary from year to year (Table 1). Hence, for every year a TF has been calculated with the realization that no steady state is achieved. In the calculations it has been assumed that the transport time from (S) to the Cattegat is approx. 4 years[3], while it takes 2 years from the waterborne radioactivity to go from (H) to the Cattegat and one year to the German Bight[4].

Four stations in the southern Cattegat were selected[3]: Kullen: 56°15'N, 12°25'E; Hesselø: 56°10'N, 11°47'E; Odden: 56°07'N, 11°10'E and Asnæs: 55°38'N, 10°47'E. Fifty-liter samples of surface and bottom water were collected summer and winter from these stations. All samples were analysed for $^{137}Cs$, and $^{90}Sr$ was determined in about 40% of the samples (Table 2).

As it appears from Table 1, the discharges from (H) are generally lower than those from (S). This is in particular the case for $^{137}Cs$. Hence, at most places in the North Sea it is difficult to identify signals from (H) in that those from (S) drown them. Furthermore, there is a background of old global fallout which contains $^{90}Sr$ as well as $^{137}Cs$, but is free of $^{134}Cs$.

From measured (fallout-corrected) seawater concentrations in the German Bight of $^{90}Sr$, $^{134}Cs$ and $^{137}Cs$ (Table 3) and from the radionuclide ratios in the discharge from (H) one year earlier and from (S) 3 years earlier (cf. Table 1) it is possible to estimate the relative contribution from (H) to the measured concentrations in the German Bight. The following mean values ($\pm$1 S.E.) were obtained for 1983, 1984 and 1985: $^{90}Sr$: (75$\pm$23)% $^{134}Cs$: (56$\pm$12)% and $^{137}Cs$: (65$\pm$17)%.

The correction for old global fall-out $^{90}Sr$ and $^{137}Cs$ was made by means of two relations derived from studies in the Danish Straits[5]:

Fallout $^{137}Cs$ Bq $m^{-3}$ = 18.5-0.44·salinity in o/oo

Fallout $^{90}Sr$ Bq $m^{-3}$ = 23.7-0.64·salinity in o/oo

The unit in the TF calculations will be Bq $m^{-3}$ per TBq $yr^{-1}$. This unit corresponds to µg $m^{-3}$ per ton $yr^{-1}$ (1 TBq = $10^{12}$ Bq).

In the data treatment, a computer program VAR-3[6] was applied. A 3-sided analysis of variance is carried out, in which missing data are calculated by assuming that no interactions are present among the parameters. The parameters were: year (1980-1985), season (summer, winter) and location (Kullen, Hesselø, Sj. Odde, Asnæs). Six analysis of variance (anova) were carried out: $^{90}Sr$ surface water, $^{90}Sr$ bottom water, $^{134}Cs$ surface, $^{134}Cs$ bottom, $^{137}Cs$ surface and $^{137}Cs$ bottom. From these anovas annual mean values for Cattegat water was calculated for each of the 3 radionuclides by combining the values for summer and winter, bottom and surface and the four stations (cf. Table 4). A similar calculation was made for bottom water only (see also Table 4).

In the subsequent calculations of transfer factors, observations covering all six years were used for $^{90}Sr$ because the discharges from (H) in all years have been significant compared with those from (S). In case of $^{134}Cs$ and $^{137}Cs$ data of the last three years (1983-1985) only were applied, because in this period the signal-to-noise ratio between (H) and (S) was 3 times better than for the period 1980-1982. In general, the signal-to-noise ratio for $^{90}Sr$ (1980-1985) was twice as good as for $^{134}Cs$, and 9 times better than for $^{137}Cs$ (both 1983-1985).

## RESULTS AND DISCUSSION

The anovas showed that there were no significant interactions among the 3 parameters (year, season and location). Hence it was valid to apply the VAR-3 programme to fill in missing values and calculate annual mean values. The anovas furthermore showed that in case of $^{137}Cs$ there was a significant difference between the years, the levels were decreasing from 1980 to 1985. The bottom water showed higher $^{137}Cs$ concentrations during the summer than in winter, while the opposite was the case for the surface water. There was no significant difference among the four locations with respect to $^{137}Cs$ levels. Surface water also showed higher $^{134}Cs$ concentrations during winter than in summer. Furthermore the bottom water from Kullen contained higher $^{134}Cs$ levels than bottom water from Asnæs. The $^{90}Sr$ data showed significant main effects. In the anovas an effect was considered significant if the significance level: $P > 99\%$.

The fallout-corrected concentrations derived from VAR-3 shown in Table 4 are due to two sources: (S) and (H). Each source has a transfer factor. In the calculation of the most likely transfer factor for (H) the following method was used: The transfer factor for (S) was varied and for each value the mean transfer factors for (H) was calculated for each of the 3 radionuclides. Then the mean of the 3 means was calculated. The transfer factor from (S) that gave the lowest relative standard deviation

of the mean for (H) of the 3 radionuclides was selected as the most likely value.

For a transfer factor from Sellafield to Cattegat of 0.015 Bq $m^{-3}$ per TBq $yr^{-1}$, the following transfer factors from Cap de la Hague to Cattegat were obtained:

$^{90}Sr$: 0.125 Bq $m^{-3}$ per TBq $yr^{-1}$ (1980-1985)
$^{134}Cs$: 0.136 ------ " ------ (1983-1985)
$^{137}Cs$: 0.119 ------ " ------ (1983-1985)

The mean of these factors were 0.127 Bq $m^{-3}$ pr TBq $yr^{-1}$ and the relative S.D. was 6.8%.

If the transfer factor for Sellafield had been changed downward to 0.014 the relative S.D. had increased to 18.6% and an upward change to 0.016 had resulted in a relative S.D. of 45.2%.

In a similar way we may calculate the transfer factor to bottom water in Cattegat. The annual mean values shown in Table 4 were used and the least relative standard deviation (22%) between the La Hague transfer factors for the 3 radionuclides was obtained for a transfer factor for Sellafield effluent of 0.022 Bq $m^{-3}$ per TBq $yr^{-1}$. The mean transfer factor from La Hague to Cattegat bottom water became 0.15 Bq $m^{-3}$ per TBq $yr^{-1}$.

From Tables 3 and 1 the transfer factors from La Hague to the German Bight were calculated, assuming a transport time of 1 year. In the calculation the above given percentages for La Hague contribution to the activity in the German Bight were applied:

$^{90}Sr$: 0.47·0.75 = 0.35 Bq $m^{-3}$ per TBq $yr^{-1}$ (1979-1985)
$^{134}Cs$: 0.57·0.56 = 0.32 ------ " ------ ( " " )
$^{137}Cs$: 0.65·0.65 = 0.42 ------ " ------ ( " " )

The mean transfer factor from La Hague to the German Bight becomes 0.36 Bq $m^{-3}$ pr TBq $yr^{-1}$. The relative standard deviation was 14%.

It is now possible to calculate the ratio of the transfer factors to the Cattegat and the German Bight and thus estimate what the concentration of a waterborne element from the German Bight may be when it shows up in the Cattegat. This calculation is valid only provided the activity from La Hague which is observed in the Cattegat, all has passed through the German Bight.

The ratio of the concentration found in total Cattegat water and to that in German Bight water becomes 0.35±0.06 (1 S.D.), and for bottom water the ratio becomes 0.42±0.11 (1 S.D.). In other words, about 40% of the bottom water in the southern part of the Cattegat may come from the German Bight.

In order to have an independent check on the above calculations it is possible to use $^{99}Tc$. As seen in Table 1, in the later years this radionuclide has been discharged in significantly larger quantities from La Hague than from Sellafield. In 1985 the expected concentration in Cattegat from La Hague would have been (0.127±0.009)·11.7 = 1.49 ± 0.11 (1 S.D.) Bq $^{99}Tc$ $m^{-3}$. The measured mean level at a coastal position Klint[3] (55°58'N, 11°35°E) in southern Cattegat was 1.06±0.55 (1 S.D; N = 9). In the Jutland Current (56°00'N, 8°03'E and 56°00'N, 7°05'E) the mean concentration was 5.0 ± 0.85 Bq $^{99}Tc$ $m^{-3}$ (1 S.D., N = 2). This activity probably represents the 1984 rather than 1983 discharge. Unfortunately we have no information on the $^{99}Tc$ from la Hague in 1984, but we assume it to be between the discharges in 1983 and 1985, i.e. 12-25 TBq. This would correspond to a transfer factor from La Hague to the Jutland Current of 0.2-0.4 Bq $m^{-3}$ per TBq $år^{-1}$, which agrees with the transfer factors calculated for the German Bight.

## CONCLUSION AND FUTURE PLANS

By means of radioactive tracers discharged from the French reprocessing plant Cap de la Hague, it is shown that about one third

of the water in the Cattegat may come from the German Bight. It further appears that about 40% of the bottom water may come from the German Bight.

By means of $^{99}Tc$ it should be possible in the coming years to verify this presumption. After the Chernobyl accident radiocaesium can no longer be used for such a study, but $^{99}Tc$ was not produced to any significant extent by the Chernobyl accident[7]. Furthermore, the fallout background and the contribution of $^{99}Tc$ from Sellafield are now both low compared to the signal from La Hague.

A 3-year project has been proposed to the Danish authorities, which implies collection and $^{99}Tc$ analysis of about 135 samples annually.

The samples will consist of:
- monthly seawater samples collected from two stations in the German Bight and two in the Cattegat
- water samples from ship cruises to the Cattegat and in the Jutland Current
- monthly seaweed samples from a coastal location in southern Cattegat
- quarterly water samples from a station, which monitors the outlet from the Baltic Sea.

The aim is to improve the calculations of the transfer of water from the German Bight to the Cattegat and to elucidate the variations in this transfer. The estimated costs of the project is about 300,000 ECU.

Table 1. Annual discharges of $^{90}Sr$, $^{134}Cs$, $^{137}Cs$ and $^{99}Tc$ from Sellafield[1] and Cap de la Hague[2]

TBq $yr^{-1}$ decay corrected to 1983

| Sellafield | 1976 | 1977 | 1978 | 1979 | 1980 | 1981 | 1982 | 1983 | 1984 | 1985 |
|---|---|---|---|---|---|---|---|---|---|---|
| $^{90}Sr$ | 324 | 370 | 531 | 229 | 328 | 267 | 311 | 204 | 74 | 55 |
| $^{134}Cs$ | 70 | 79 | 75 | 61 | 87 | 87 | 99 | 89 | 49 | 59 |
| $^{137}Cs$ | 3649 | 3898 | 3641 | 2336 | 2767 | 2292 | 1954 | 1200 | 444 | 340 |
| $^{99}Tc$ | ? | ? | 179 | 43 | 57 | 5,8 | 3,6 | 4,4 | 4,3 | 1,9 |

| Cap de la Hague | 1978 | 1979 | 1980 | 1981 | 1982 | 1983 | 1984 | 1985 | 1986 | 1987 |
|---|---|---|---|---|---|---|---|---|---|---|
| $^{90}Sr$ | 62 | 53 | 27,4 | 25,8 | 84 | 142 | 112 | 49 | | |
| $^{134}Cs$ | 1,46 | 0,93 | 1,43 | 3,04 | 6,01 | 4,92 | 6,73 | 16,0 | | |
| $^{137}Cs$ | 34,78 | 20,54 | 24,97 | 36,85 | 49,38 | 23 | 31 | 31 | | |
| $^{99}Tc$ | ? | ? | ? | ? | ? | 11,7 | ? | 25,4 | 9,8 | |

Table 2. Sr-90, Cs-134 and Cs-137 concentrations (Bq $m^{-3}$) at four stations in the southern Cattegat[3)] in surface water (OV) and bottom water (BU) in the period 1980-1985. (All data are decay corrected to July 1, 1983) (S: Summer V: Winter)

| | | | Kullen Station 1 | | | Hesselø Station 2 | | | Sj. Odde Station 3 | | | Asnæs Station 4 | | | Mean salinity |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Year | Water layer | Season | $^{90}Sr$ | $^{134}Cs$ | $^{137}Cs$ | $^{90}Sr$ | $^{134}Cs$ | $^{137}Cs$ | $^{90}Sr$ | $^{134}Cs$ | $^{137}Cs$ | $^{90}Sr$ | $^{134}Cs$ | $^{137}Cs$ | o/oo |
| | OV | S | 21 | 0,58 | 38 | 23 | 0,76 | 45 | 17 | - | 35 | - | - | 33 | 15,3 |
| 1980 | OV | V | 14 | 1,21 | 63 | 10 | 1,47 | 64 | 13 | 1,03 | 47 | 20 | 1,34 | 63 | 23,0 |
| | BU | S | 22 | 1,97 | 105 | 24 | 1,67 | 99 | 22 | 1,64 | 104 | - | 2,00 | 98 | 31,8 |
| | BU | V | 13 | - | - | 22 | 2,20 | 102 | - | 1,12 | 48 | 21 | 1,81 | 90 | 31,9 |
| | OV | S | 19 | - | 33 | - | 0,92 | 48 | 17 | 1,17 | 48 | - | 1,12 | 49 | 16,6 |
| 1981 | OV | V | - | - | - | 31 | 1,63 | 72 | - | 1,75 | 66 | 25 | 1,63 | 61 | 22,7 |
| | BU | S | 26 | 1,99 | 96 | - | 1,73 | 83 | 25 | 2,09 | 93 | - | 1,84 | 83 | 33,4 |
| | BU | V | - | - | - | 20 | 1,27 | 74 | - | 1,69 | 68 | 20 | 0,91 | 53 | 22,8 |
| | OV | S | - | - | 33 | 19 | 0,94 | 29 | | - | 29 | | - | 27 | 13,4 |
| 1982 | OV | V | 20 | - | 39 | - | 1,69 | 56 | 18 | 2,03 | - | 18 | 2,11 | 59 | 22,1 |
| | BU | S | - | 1,71 | 74 | - | 1,50 | 80 | 16 | 1,71 | 72 | - | 2,00 | 71 | 32,3 |
| | BU | V | 17 | - | 56 | 17 | 2,28 | 64 | - | - | 59 | 16 | 2,11 | 61 | 26,7 |
| | OV | S | - | - | 24 | 21 | 0,82 | 45 | - | - | 31 | 21 | - | 24 | 14,8 |
| 1983 | OV | V | - | - | 23 | - | 2,13 | 52 | - | | - | - | 0,60 | 32 | 17,7 |
| | BU | S | 19 | 2,70 | 74 | - | 2,40 | 60 | 15 | 2,40 | 66 | - | 1,60 | 67 | 32,7 |
| | BU | V | - | 2,13 | 60 | 21 | 1,15 | 61 | - | - | - | 23 | 0,71 | 45 | 25,7 |
| | OV | S | - | - | 27 | - | - | 30 | - | - | 33 | - | - | 26 | 13,9 |
| 1984 | OV | V | 12 | - | 46 | - | 2,65 | 55 | - | - | 37 | - | - | 41 | 20,0 |
| | BU | S | - | 2,94 | 90 | - | 2,62 | 89 | 25 | 2,10 | 83 | - | 2,28 | 76 | 33,6 |
| | BU | V | - | 4,64 | 75 | - | 2,62 | 67 | 29 | - | 46 | - | 1,99 | 55 | 30,6 |
| | OV | S | - | - | 32 | 19,7 | 0,98 | 39 | - | 0,63 | 25 | - | 0,36 | 22 | 13,7 |
| 1985 | OV | V | - | - | 25 | - | - | 31 | 23 | - | 40 | - | - | 30 | 15,9 |
| | BU | S | - | 2,63 | 79 | - | 2,32 | 72 | - | 2,58 | 78 | 22 | 2,00 | 67 | 33,0 |
| | BU | V | 27 | 2,19 | 53 | - | 2,65 | 50 | - | 1,57 | 51 | - | 1,10 | 44 | 30,5 |

Table 3. Concentrations of $^{90}Sr$, $^{134}Cs$ and $^{137}Cs$ in the German Bight (Bq $m^{-3}$). Decay corrected to July 1, 1983)

| Year | $^{90}Sr$ | $^{134}Cs$ | $^{137}Cs$ |
|---|---|---|---|
| 1979 | 26,4 | 0,62 | 18,8 |
| 1980 | 16,8 | 0,74 | 16,8 |
| 1981 | 16,2 | 1,28 | 22,9 |
| 1982 | 16,6 | 1,80 | 22,5 |
| 1983 | 37 | 1,96 | 16,5 |
| 1984 | 56 | 3,71 | 19 |
| 1985 | 45 | 3,31 | 15 |

The figures are based upon Kautsky's data[4] and Risø measurement[3]; $^{134}Cs$ and $^{137}Cs$ were either measured directly or calculated from the sum of the two isotopes and from their ratio in the Cap de la Hague discharges one year earlier (cf. Table 1).

Table 4. VAR 3[6] calculated mean concentrations (Bq $m^{-3}$) in the southern Cattegat in the period 1980-1985. Sr-90 and Cs-137 are corrected for fallout background (cf. the text). All data are decay corrected to July 1, 1983.

| Year | $^{90}Sr$ | | $^{134}Cs$ | | $^{137}Cs$ | |
|---|---|---|---|---|---|---|
| | total | bottom | total | bottom | total | bottom |
| 1980 | 11.7 | 17.3 | 1.36 | 1.78 | 62.7 | 86.9 |
| 1981 | 14.5 | 17.1 | 1.53 | 1.69 | 58.1 | 71.8 |
| 1982 | 9.2 | 11.9 | 1.69 | 1.91 | 45.3 | 61.6 |
| 1983 | 10.5 | 14.5 | 1.43 | 1.84 | 38.4 | 54.2 |
| 1984 | 12.0 | 24.7 | 2.44 | 2.71 | 47.1 | 68.2 |
| 1985 | 13.2 | 18.7 | 1.57 | 2.13 | 37.9 | 57.3 |

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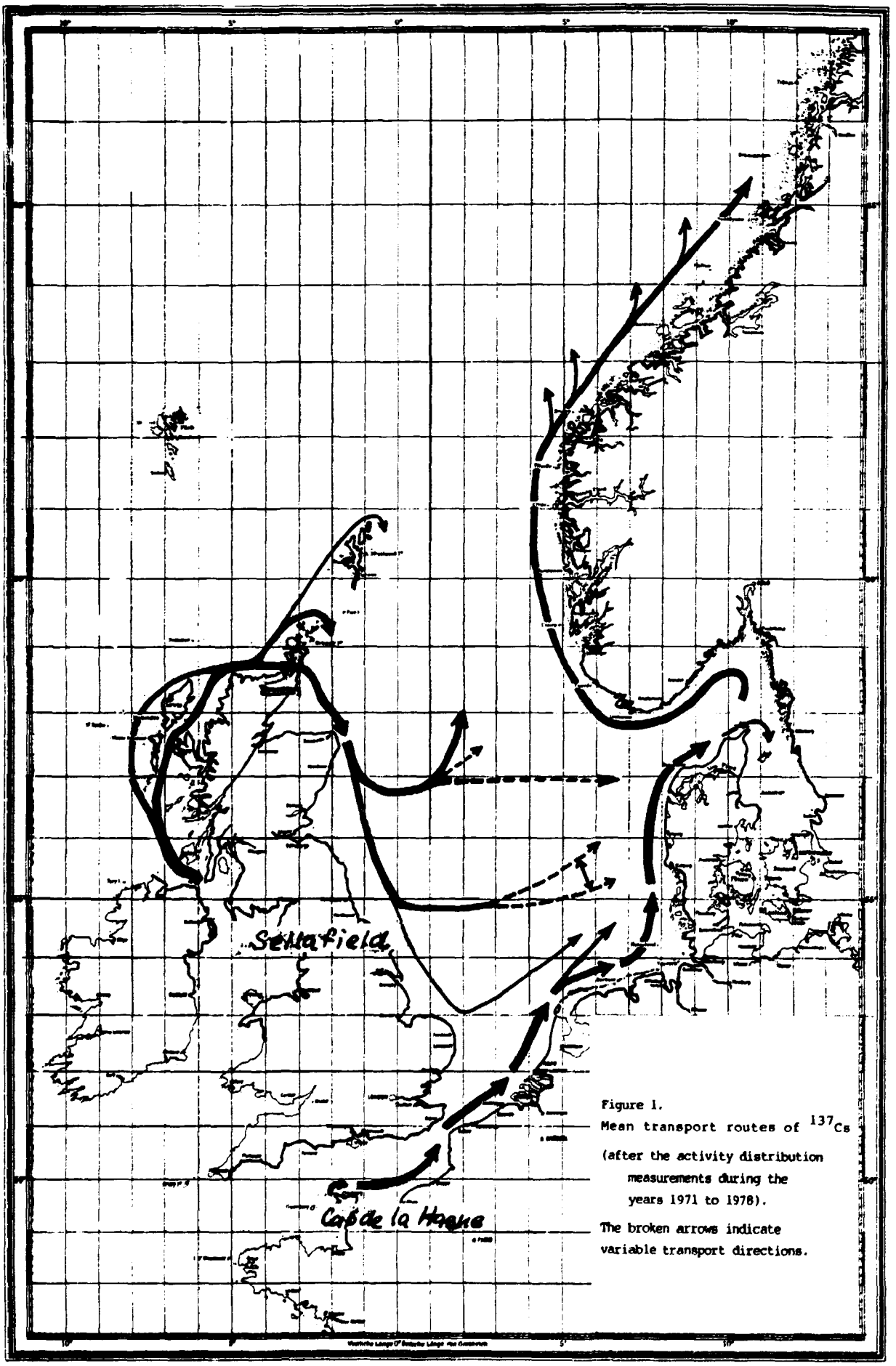


Figure 1.
Mean transport routes of $^{137}Cs$
(after the activity distribution measurements during the years 1971 to 1978).
The broken arrows indicate variable transport directions.

(After H. Kautsky)

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