Iodine as a geotracer

Sources of ^{129}I to the ocean

Natural

- spallation of Xe in the atmosphere ~80-100 kg
- fission of $^{238,235}U$ in the earth's crust

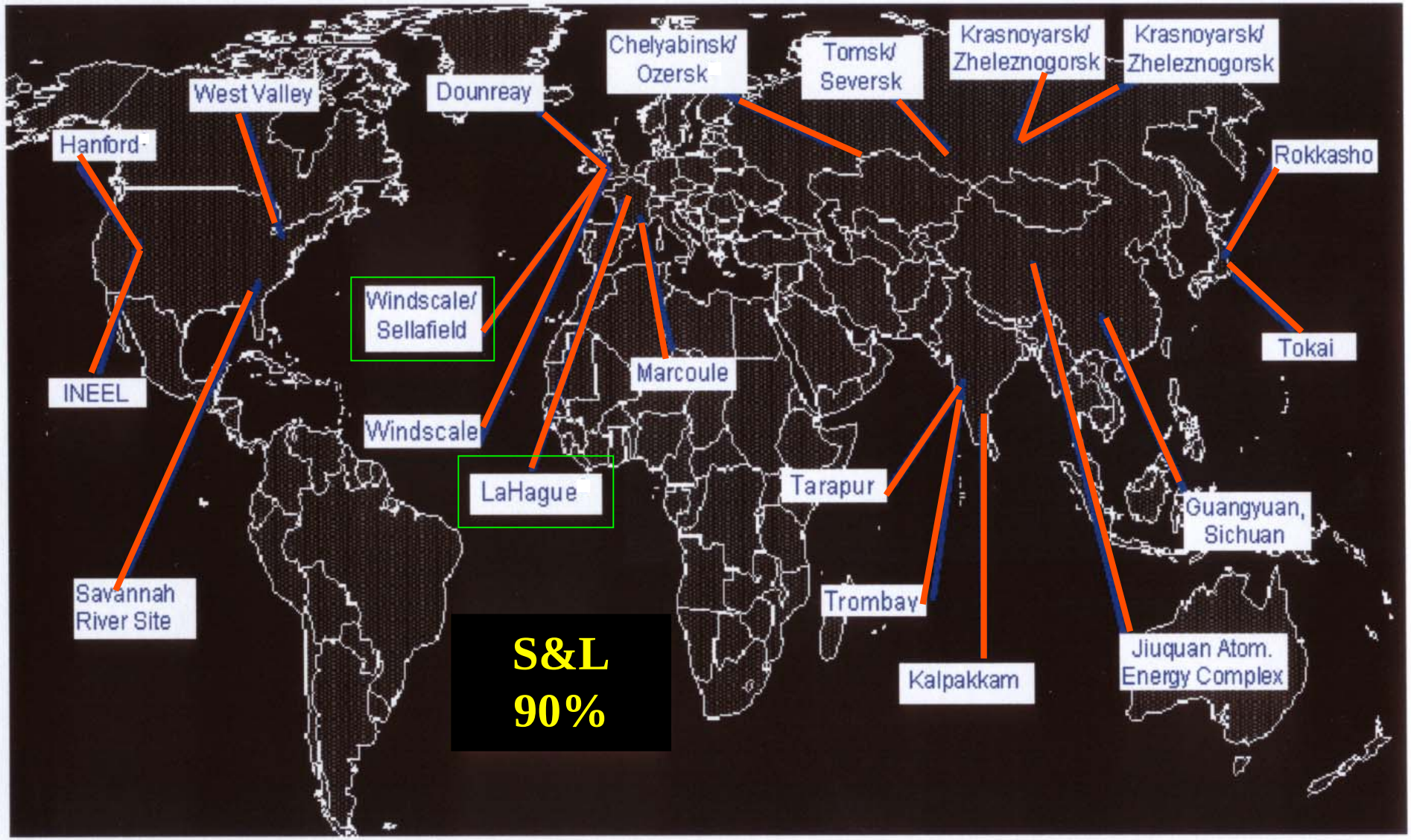
(Natural ocean water $^{129}I/^{127}I \sim 10^{-12}$)

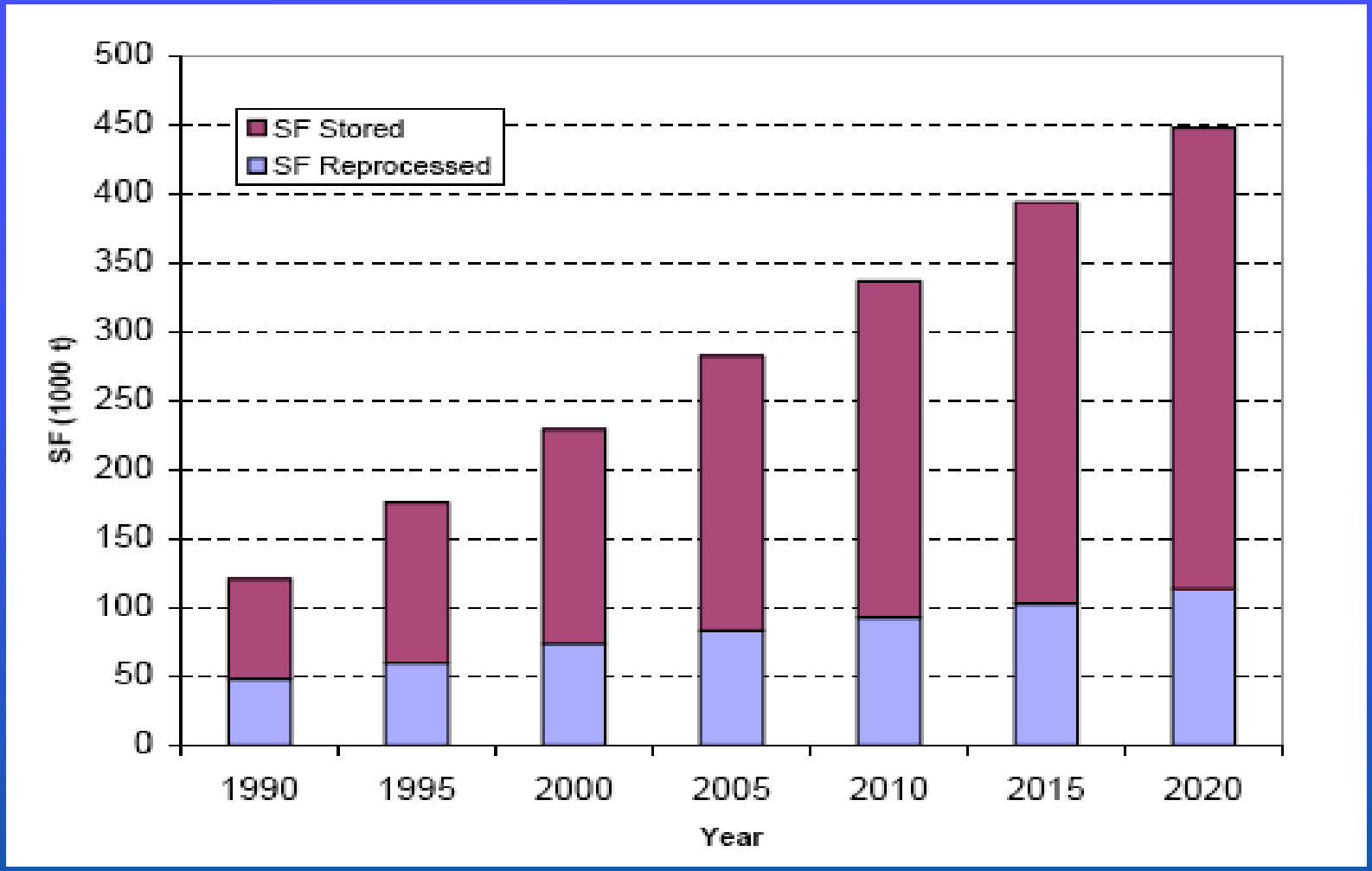
Anthropogenic

- Nuclear bomb testing (1952-1963) ~ 50 kg
- Reactor accidents (e.g. Chernobyl, 1986) ~ ? kg
- Nuclear fuel reprocessing ~ 5000? kg

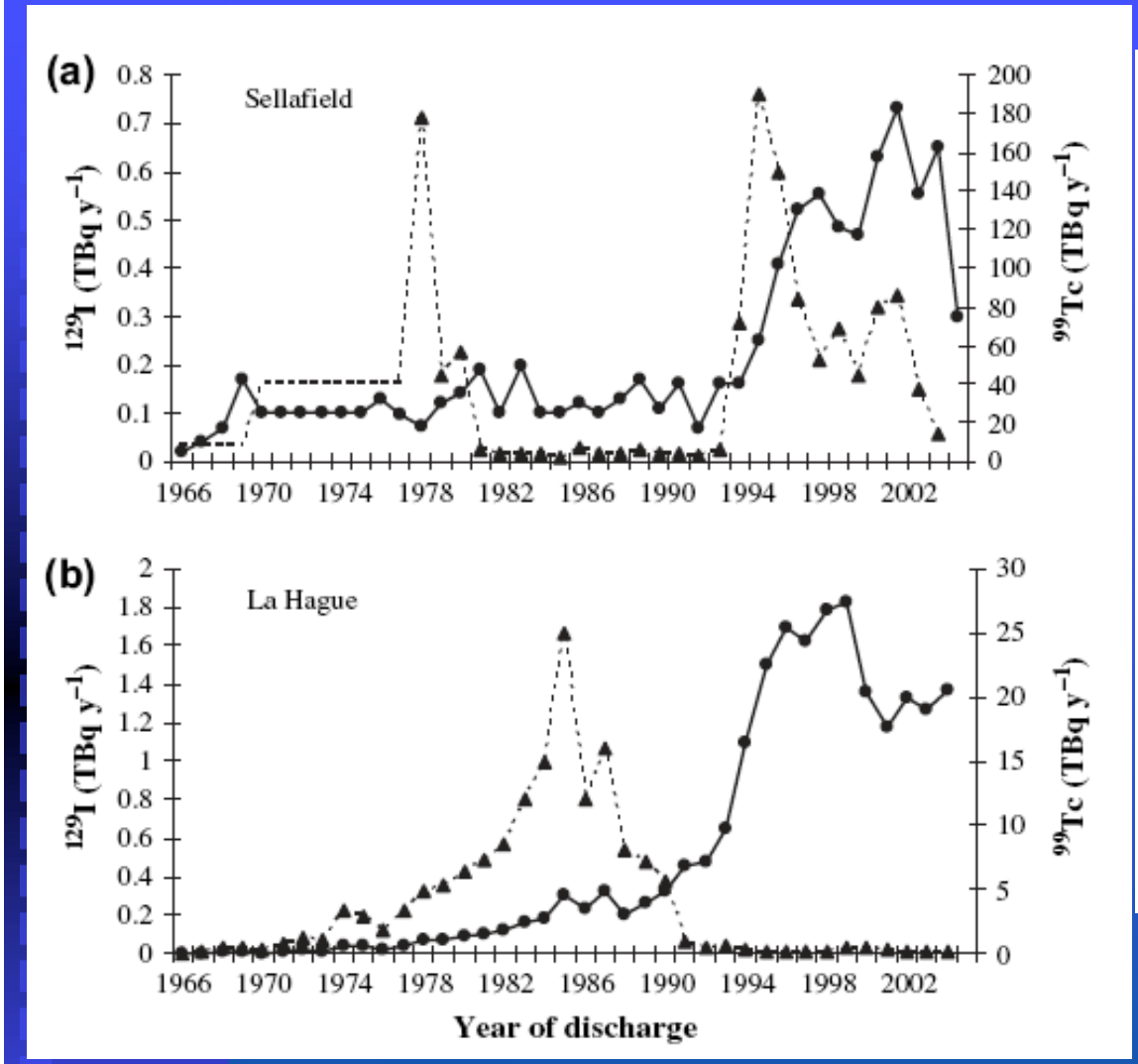
(Anthropogenic ocean water $^{129}I/^{127}I \sim 10^{-10}$ -10^{-6})

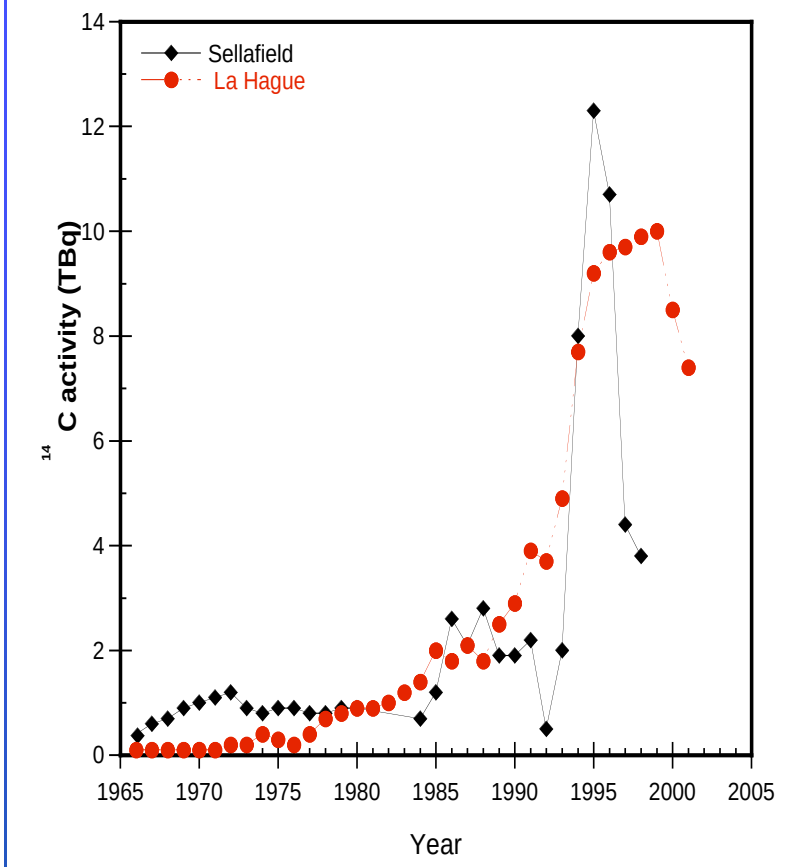
Nuclear Fuel Reprocessing Sites in the World





Trends in the global spent fuel development
(IAEA-TECDOC-1467, 2005)





Liquid emissions from Sellafield (a) and La Hague (b), ^{129}I (solid line) and ^{99}Tc (dotted line) (Keogh et al., 2007), and ^{14}C emission (c) (Gulliver et al., 2004 and Douvelle et al., 2004).

Questions:

What is the input function ?
No historical marine archive available yet

How conservative is iodine?
(depends on sedimentation rates)

What are the pathways?

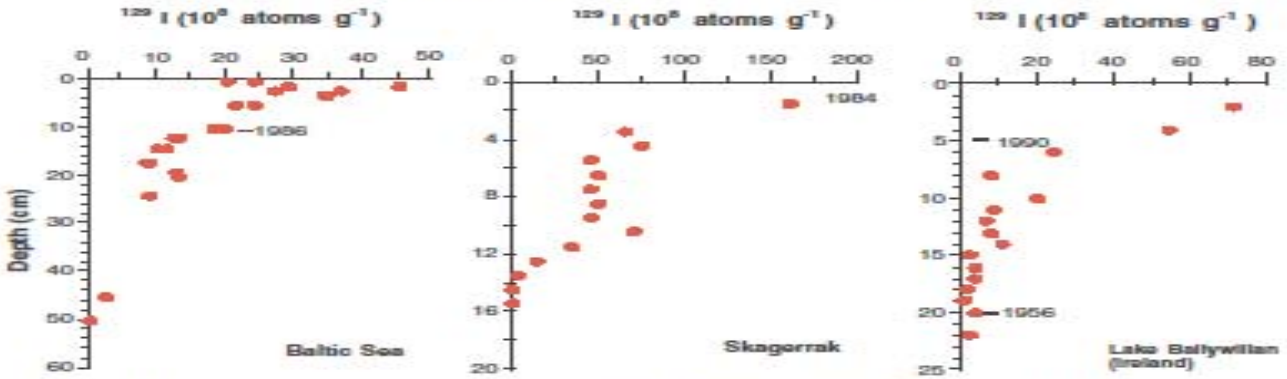


Fig. 8. Distribution of ^{129}I in the studied sediment, Skagerrak (López-Gutiérrez et al., 2004) and Lake Ballywillan (Ireland, Gallagher et al., 2005).

Replenishment of the Baltic Sea water by the saline North Sea water via the Skagerrak and Kategatt basins is generally sporadic (Meier, 2005 and references therein). Available data on ^{129}I from the water column of the these two basins indicate concentrations about 1–2 orders of magnitude higher than in the Baltic Sea and also a clear gradient of decreasing concentrations towards the northern parts of the Baltic Sea (Aldahan et al., 2006). Presently, there are no data on the ^{129}I and ^{127}I contents of river sediments which renders it difficult to estimate the terrestrial I contribution in the Baltic Sea sediment. Investigation of ^{129}I in river waters of the Baltic region suggests that river water inflow and atmospheric transport represents a relatively small amount (<1%) of the total inventory in the Baltic Sea (Aldahan et al., 2005). This, together with ^{129}I concentrations that are an order of magnitude lower (10^8 atoms/g) in the top 10 cm of soils in central Sweden (Hou et al., 2003; Englund et al., 2006) than in the studied cores, indicates that the supply of ^{129}I by detritus can be considered small and most likely uniform over the last 100 a. These observations and the relatively small (<10%) contribution from the Chernobyl accident (Aldahan et al., 2006) suggest that most of the ^{129}I in the Baltic Sea can be related to the liquid discharges from the Sellafield and La Hague nuclear reprocessing facilities.

Estimated transit time for the ^{129}I plume of Sellafield to reach the North Sea is about 1.5–2 a and that of the La Hague is 0.5–1 a (Bailly du Bois et al., 1995; Kershaw and Baxter, 1995; Hou et al., 2002). Additional time is needed for the combined plume to reach the studied sites, although the generally sporadic nature of currents entering from the Baltic Sea from the Kategatt Basin and the variable residence time within the Baltic Sea makes this estimate difficult (Gustafsson and Andersson, 2001; Meier, 2005). However, in order to provide a modeled comparison of the expected trend of ^{129}I liquid releases that enter the Baltic Sea, the authors have delayed the Sellafield plume to the North Sea by

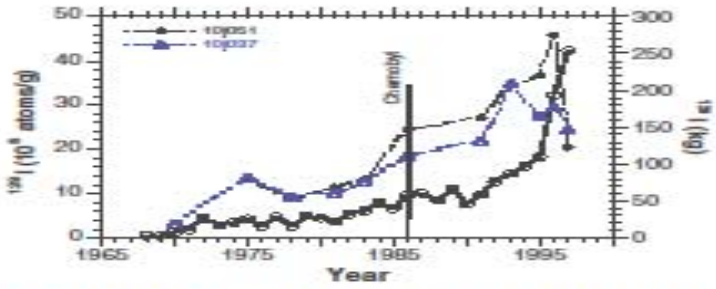
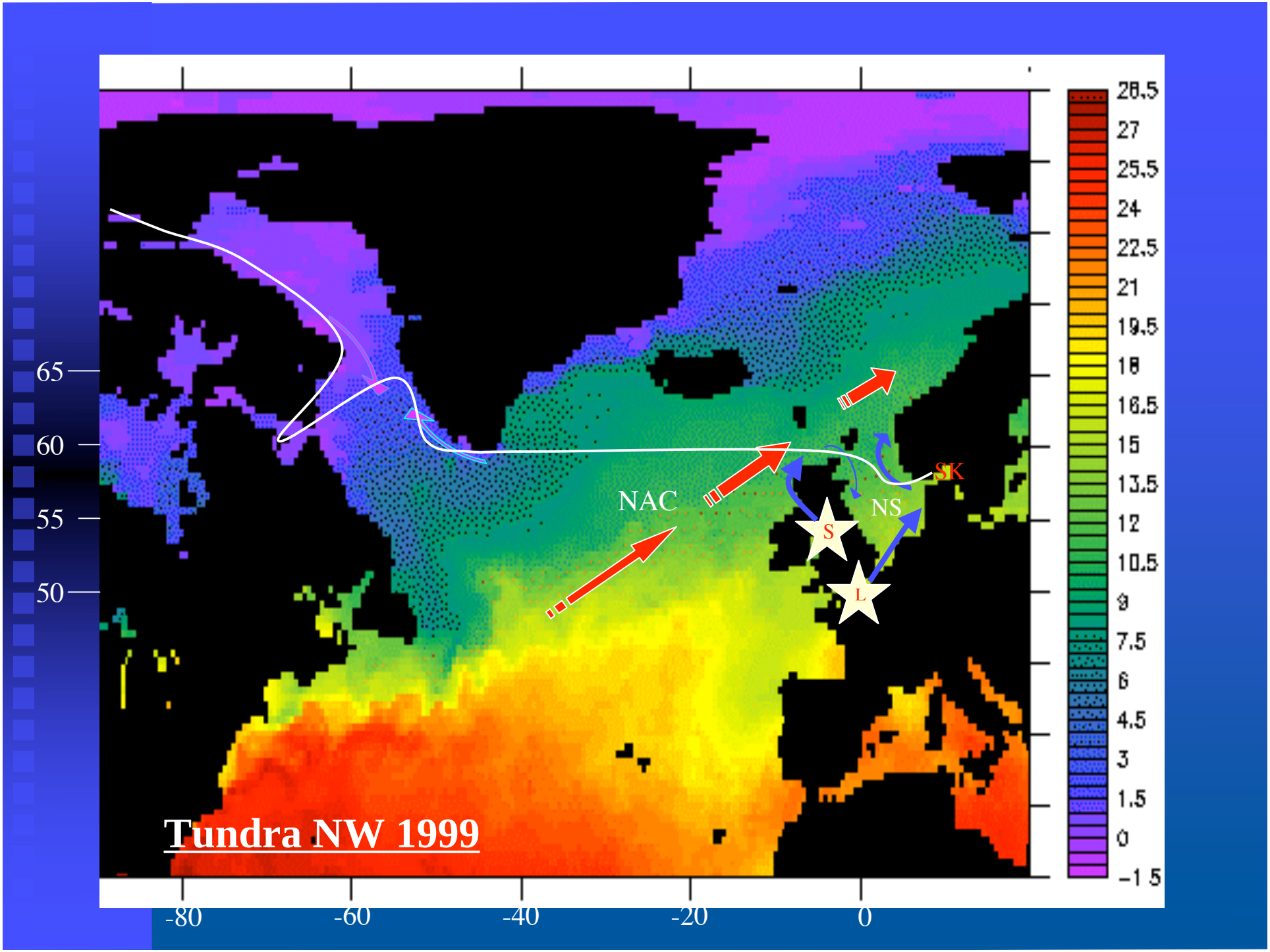
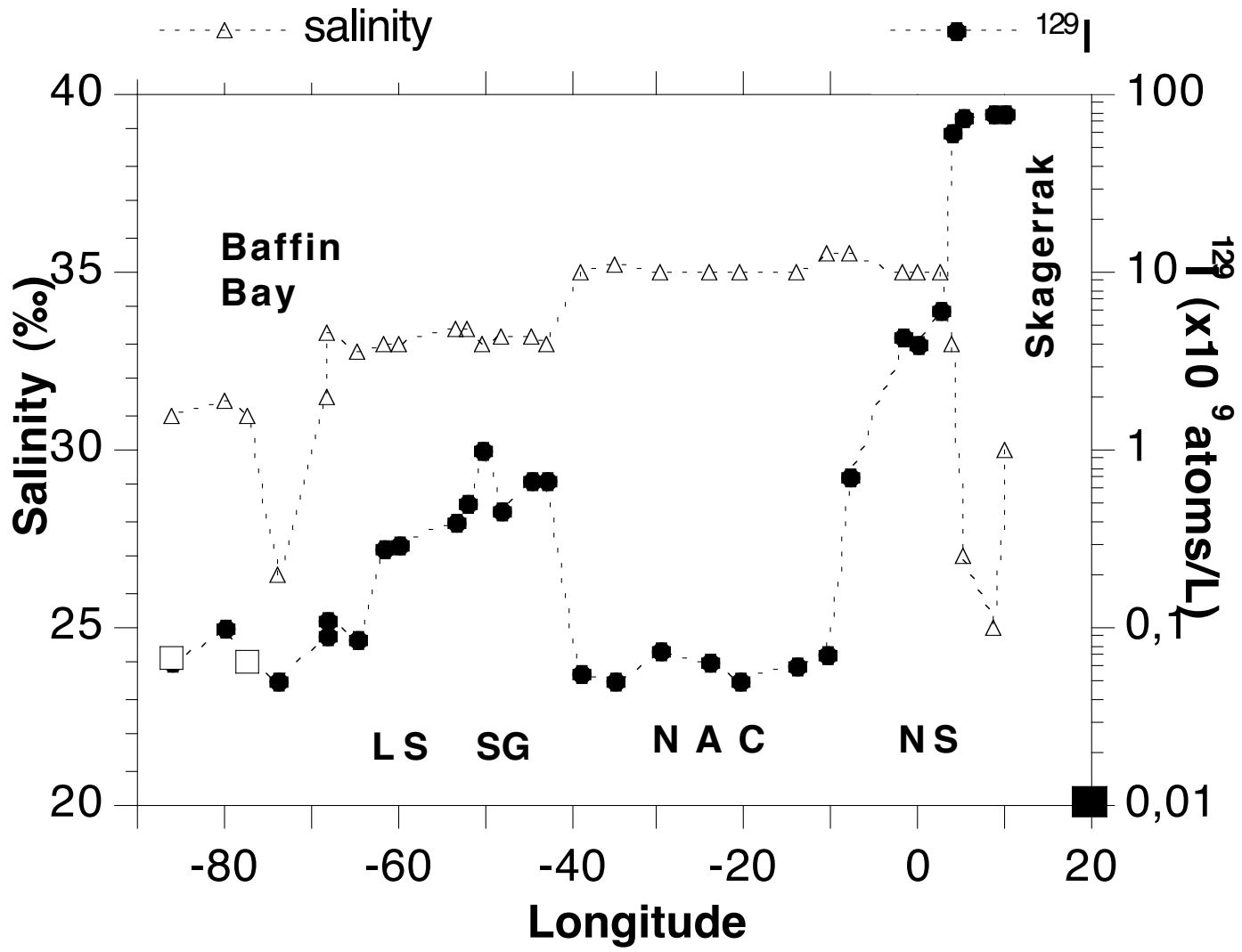
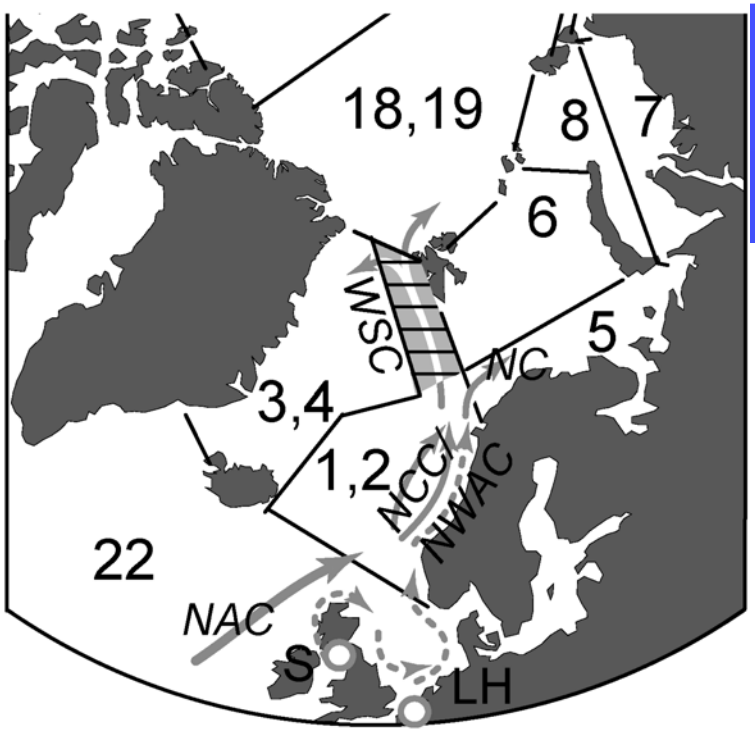


Fig. 9. Distribution of ^{129}I in the sediment cores together with total liquid releases from the Sellafield and La Hague facilities. The Sellafiled plume was delayed by one year to reach the North Sea and combined with the La Hague plume and the total was delayed by two years to reach the study area from the North Sea. Data for Sellafield and La Hague liquid discharges are from López-Gutiérrez et al. (2004).

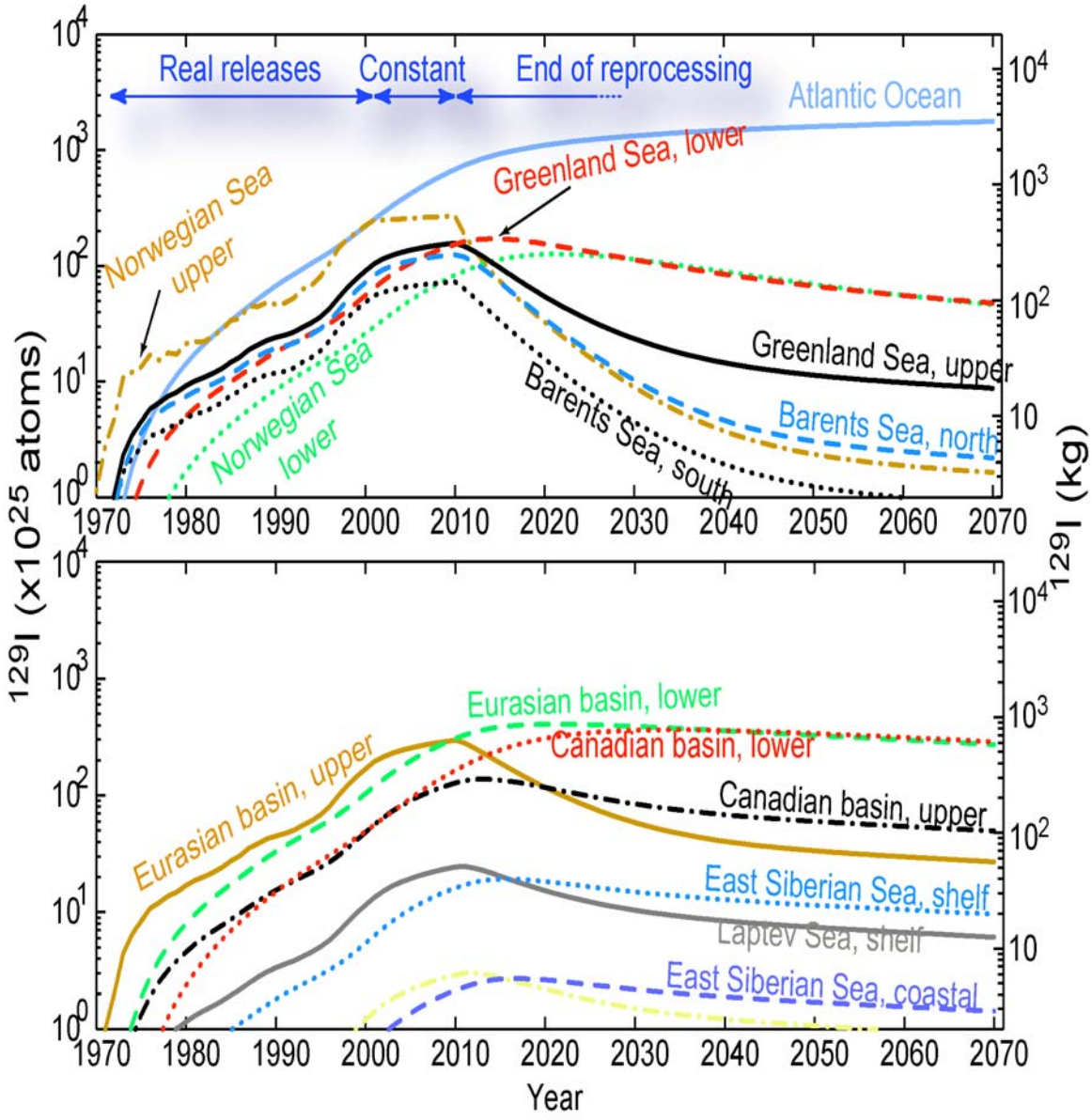

Tundra NW 1999
NAC
NS
SK
S
L
65
60
55
50
-80
-60
-40
-20
0
28.5
27
25.5
24
22.5
21
19.5
18
16.5
15
13.5
12
10.5
9
7.5
6
4.5
3
1.5
0
-1.5


salinity
129I
40
35
30
25
20
100
10
1
0,1
0,01
Salinity (‰)
129I (x10 9 atoms/L)
Baffin Bay
Skagerrak
L S
SG
N A C
N S
-80
-60
-40
-20
0
20
Longitude



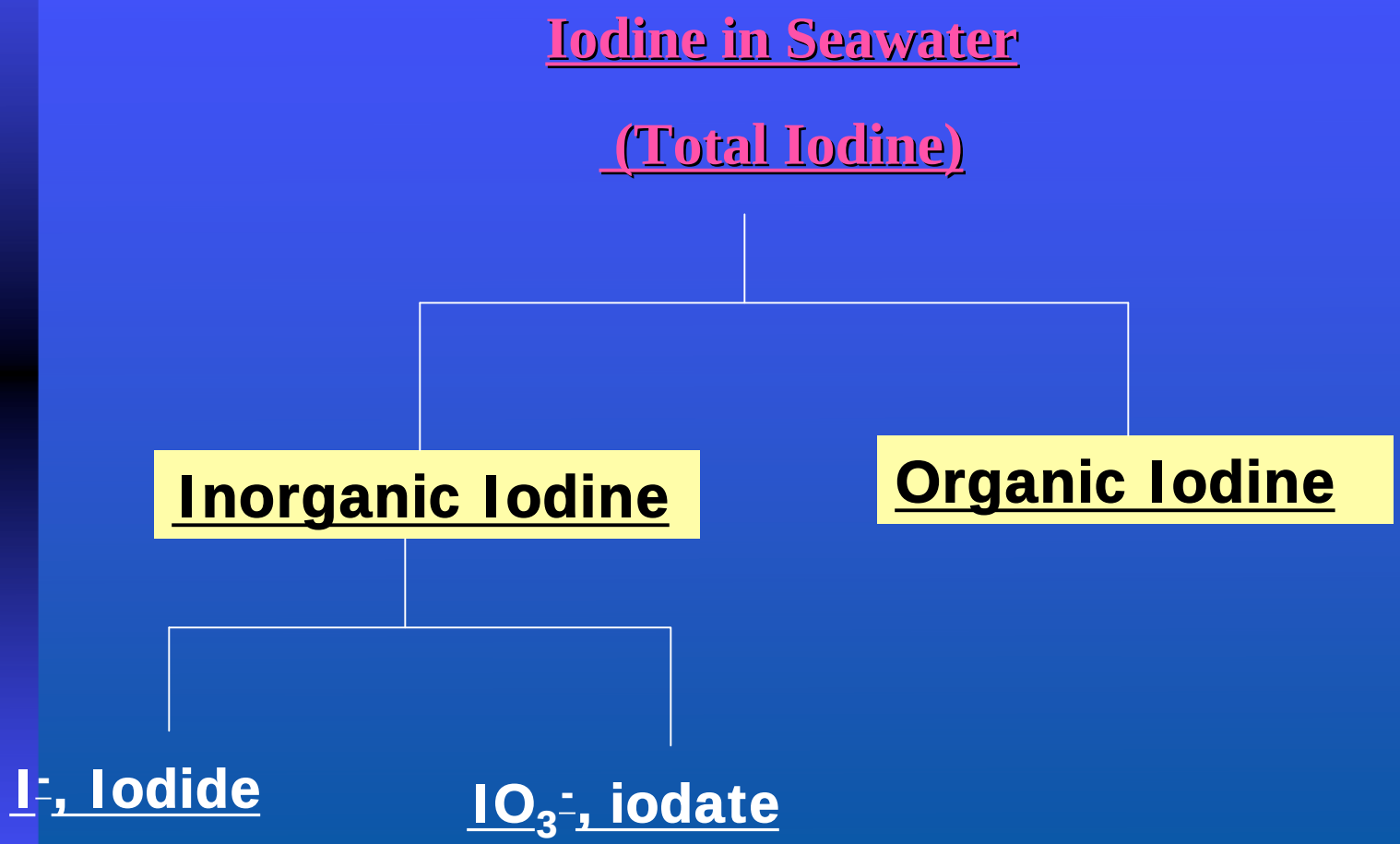
Modeling ^{129}I transport

Modified model of Risk Assessment Integration Group (Arctic Nuclear Waste Assessment Program)



Fast transport rates to and large inventories in the deep Arctic Ocean and North Atlantic

Abrupt loss to North Atlantic after fictitious closure of the Reprocessing facilities


Iodine in Seawater
(Total Iodine)
Inorganic Iodine
Organic Iodine
I-, Iodide
IO3-, iodate

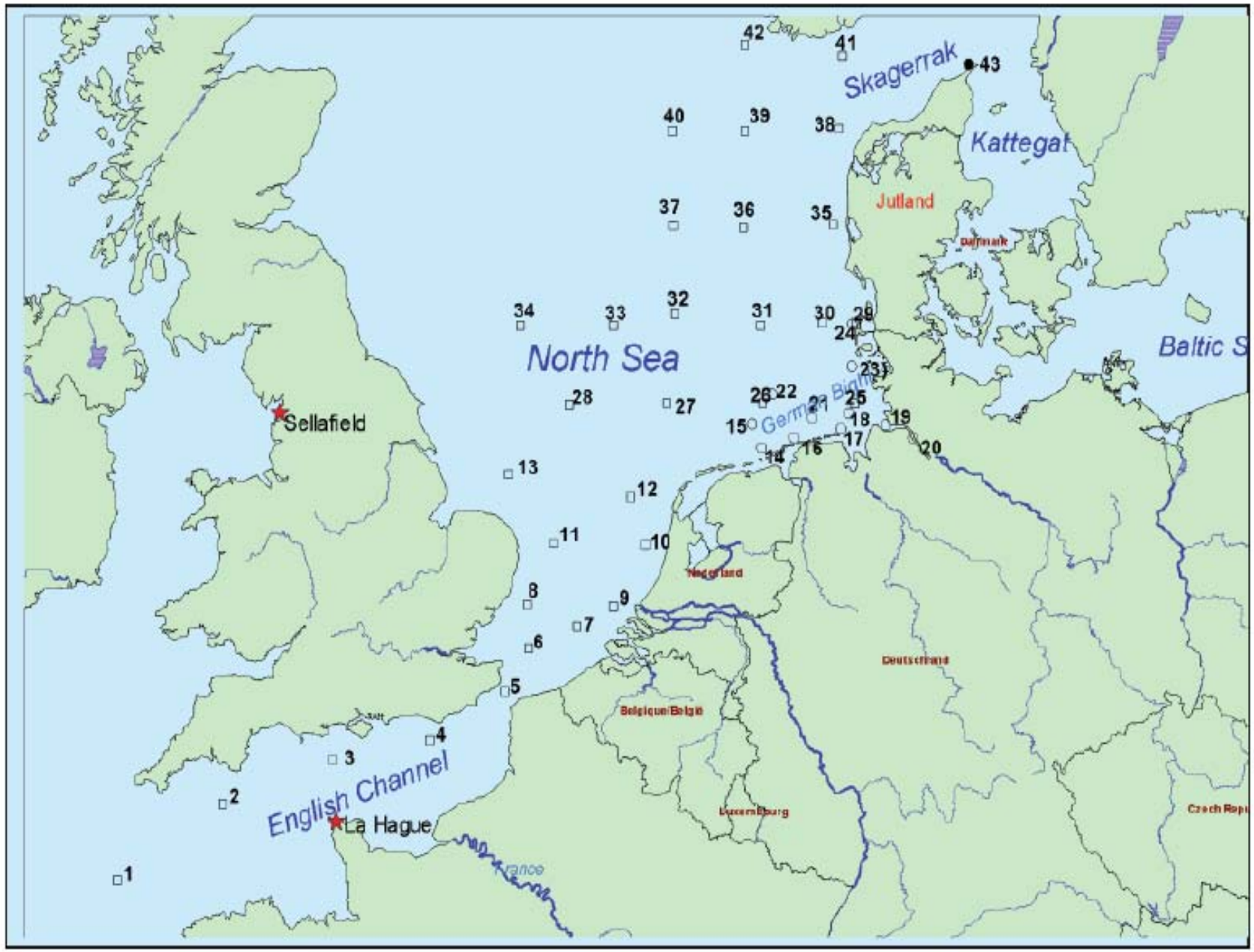


FIGURE 1. Sampling locations of surface water from the North Sea (□) and German Bight (○) in August 2005 and Danish coast in November 1999 (●).

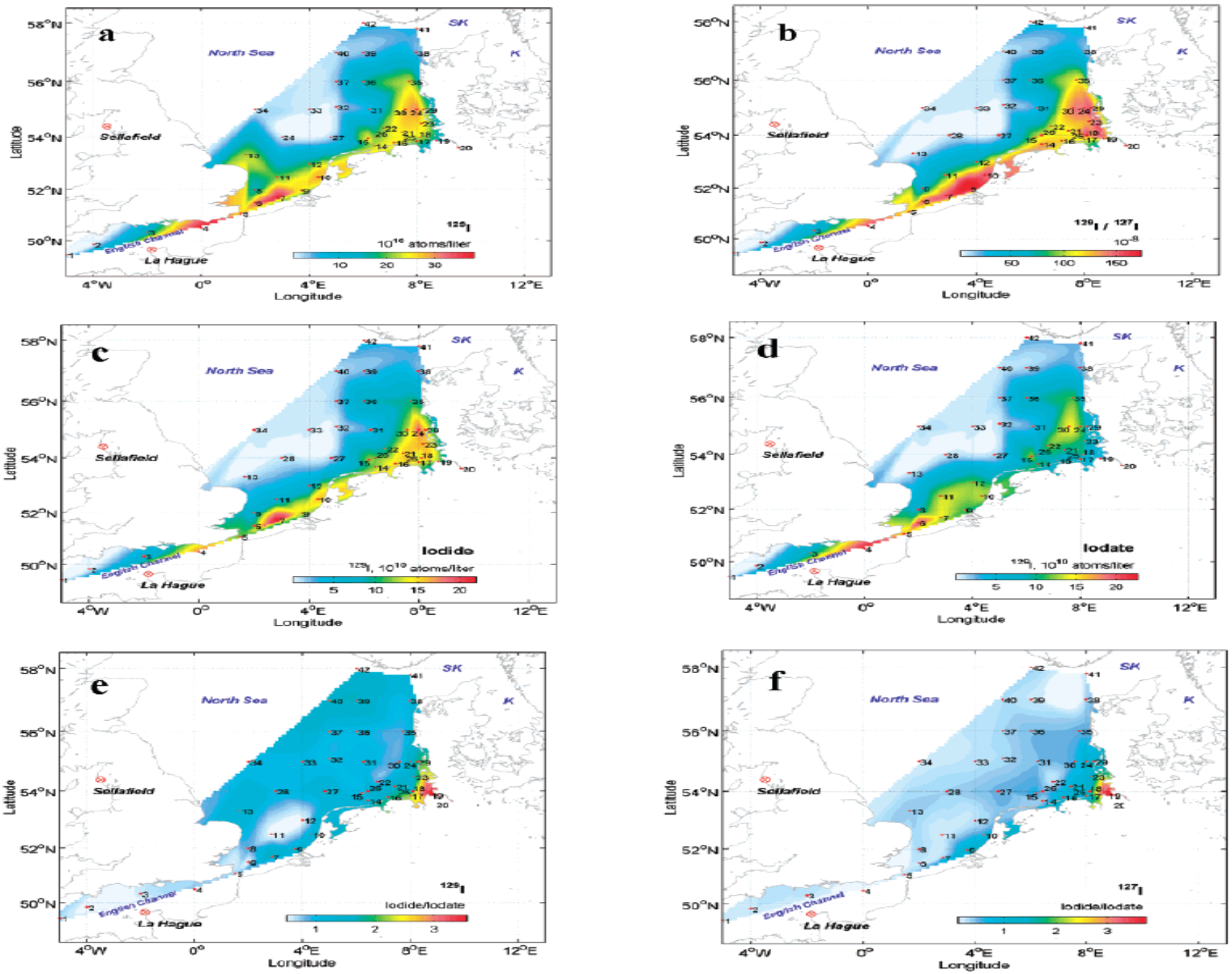


FIGURE 2. Distribution of total ^{129}I (a), ^{129}I/^{127}I atomic ratios (b), $^{129}I^-$ (c), $^{129}IO_3^-$ (d), $^{129}I^-/^{129}IO_3^-$ (e), and $^{127}I^-/^{127}IO_3^-$ molecular ratio (f) in the English Channel and the North Sea.

Speciation analysis will provide information about:

*Inflow/outflow features (rates, quantities, origin)

*Recycling

*Residence time