Available online at www.sciencedirect.com

ScienceDirect

Applied Geochemistry 22 (2007) 606–618

Applied Geochemistry

www.elsevier.com/locate/apgeochem

^{129}I anthropogenic budget: Major sources and sinks

A. Aldahan [a,*], V. Alfimov [b], G. Possnert [c]

[a] *Department of Earth Sciences, Uppsala University, SE-758 36 Uppsala, Sweden*
[b] *ETH/PSI Ion Beam Physics Group, Institute of Particle Physics, ETH Hönggerberg, 8093 Zurich, Switzerland*
[c] *Tandem Laboratory, Uppsala University, SE-751 21 Uppsala, Sweden*

Abstract

Data are presented here on the anthropogenic ^{129}I inventory in regions that have been strongly affected by releases from European reprocessing facilities which, to the authors' knowledge, presently account for >90% of the global isotope source in the Earth's surface environment. The results show that >90% of the isotope inventory occurs in marine waters with the Nordic Seas and Eurasian basin of the Arctic Ocean containing most of the ^{129}I. Within the terrestrial environment of Europe, soils contain the largest part of the isotope inventory. However, the inventory of the terrestrial system did not provide clues on the most plausible atmospheric source of ^{129}I to Europe, thus supply from both gaseous and marine releases is proposed. The sum of the total inventory in both the marine and terrestrial environments did not match the estimated releases. This imbalance is likely to relate to unconstrained inventory estimates for marine basins (Irish Sea, English Channel and North Sea) close to the facilities, but also to the occurrence of ^{129}I in the biosphere, and possible over-estimated releases from the nuclear reprocessing facilities. There is no doubt that the available data on ^{129}I distribution in the environment are far from representative and further research is urgently needed to construct a comprehensive picture.

1. Introduction

There has been considerable interest in utilizing anthropogenic ^{129}I (half-life 15.7 Ma) as a geochemical tracer in a wide range of natural reservoirs. Consequently, a large amount of data has been generated with respect to the distribution of the isotope in the hydrosphere and atmosphere and to a lesser extent in the biosphere and lithosphere. Furthermore, much of the dynamics of anthropogenic ^{129}I distribution is linked to natural ^{127}I, which may be partly misleading considering the large variability in input sources, modes and concentrations. However, as with all types of chemical tracers, conservative behavior (relatively constant concentration in a reservoir over time) is desirable in order to use the element or compound as a tracer. In the case of ^{127}I this assumption can be true, but unfortunately it is not applicable for ^{129}I as the natural concentration has been strongly overwhelmed by the continuous and sporadic additions from a variety of anthropogenic sources. This situation complicates the attempt to assign probabilities based on constant source input scenarios for ^{129}I and the consequences of providing a final assessment of global distribution and budget inventory. Accordingly, a continuously revised budget calculation is practically indispensable to trace existing and future global changes in inventories of anthropogenic ^{129}I.

---

[*] Corresponding author.
E-mail address: ala.aldahan@geo.uu.se (A. Aldahan).

0883-2927/$ - see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.apgeochem.2006.12.006

In this report, data on anthropogenic ^{129}I from the authors' group and other researchers are used to elucidate the expected concentrations and inventory in the Earth's surface environments of the Northern hemisphere and in particular Europe, the Arctic and North Atlantic Oceans and related Seas. These data are further evaluated in terms of ^{129}I mass balance through calculation of input functions and recovered inventories in the surface environments.

2. Sources of anthropogenic ^{129}I and release rates

The sources of anthropogenic ^{129}I include mainly two domains (Table 1). Those of a sporadic nature associated with nuclear weapons testing and nuclear accidents, while the other domain represents continuous releases from nuclear reprocessing facilities and nuclear power plants. Within these domains the characteristics (gaseous or liquid) and annual rate of release also vary. Furthermore, whereas clouds from most atomic weapons testing can reach the stratosphere, no such vertical atmospheric dispersal is expected by gaseous releases from other sources. The residence time of ^{129}I in the troposphere is estimated to between 2 and 4 weeks, with a likely longer period in the stratosphere. Consequently, dispersal of a stratospheric ^{129}I pulse is expected to be more global than a tropospheric one. That is also a reason for the global detection of atomic weapon related ^{129}I pulses, which have caused elevated concentration about 2 orders of magnitude above the natural values found in the Earth's surface reservoirs. In contrast to that, transport of gaseous releases from sources (e.g. reprocessing facilities) other than nuclear weapon testing would have limited temporal and spatial global spread and can take a much longer time. Thus these general guidelines provide direction for the consideration of ^{129}I distribution from release sources to the sink and corresponding inventory.

This report focuses on anthropogenic ^{129}I input from the European nuclear reprocessing facilities, which account for more than 90% of the total global releases (Table 1). There are relatively good data for the releases from the Sellafield and La Hague facilities until 2004, but less constrained data on releases from the Marcoule facility. Furthermore, the Sellafield and La Hague facilities discharge a large part of their waste directly into marine water, whereas discharge from other facilities in Table 1 (Hanford and Marcoule) was into rivers. Part of the ^{129}I in the Arctic Ocean may also be related to the releases from the Russian (Siberian) facilities, such as

Table 1
Sources of anthropogenic ^{129}I

Source	^{129}I (10^{26} atoms)	^{129}I (kg)	Release form	Reference[c]
Nuclear power reactors				
Nuclear weapons testing 1945–1970s	2–7	50–150	Gaseous	(1)–(3)
Chernobyl accident, April 1986	0.28	6	Gaseous	[a]
Nuclear reprocessing facilities				
Hanford, 1944–1972, USA[b]	12	260	Gaseous	(4)
Marcoule, 1988–1997, France[b]	3	68	Gaseous	(5)
Sellafield, 1952–2004, UK	9	182	Gaseous	(6)–(9)
	64	1371	Liquid	(7), (8)
La Hague, 1966–2004, France	3	68	Gaseous	(6), (10)
	146	3119	Liquid	(6), (10)
Others (Russia, Japan, India, China)	–	–		
Total (reprocessing)	237+	5068+		

[a] Estimated using data on ^{129}I/^{131}I from Mironov et al. (2002) and the total ^{131}I from UNSCEAR (2000).

[b] Liquid releases from Sellafield and La Hague are discharged into the sea whereas those from Hanford and Marcoule were discharged into the Columbia and Rhone rivers respectively.

[c] Reference: (1) Eisenbud and Gesell (1997), (2) Wagner et al. (1996), (3) Raisbeck and Yiou (1999), (4) Hanford (1997), (5) Cogema (1997), (6) López-Gutiérrez et al. (2004), (7) BNFL (1999-2004), (8) Gray et al. (1995), (9) Jackson et al. (2000), and (10) GRNC (1999). Presently we have no estimate for the releases from Nuclear power plants and the so called Others in the table as well as for the liquid releases from Hanford and Marcoule, which can add to the total sums.

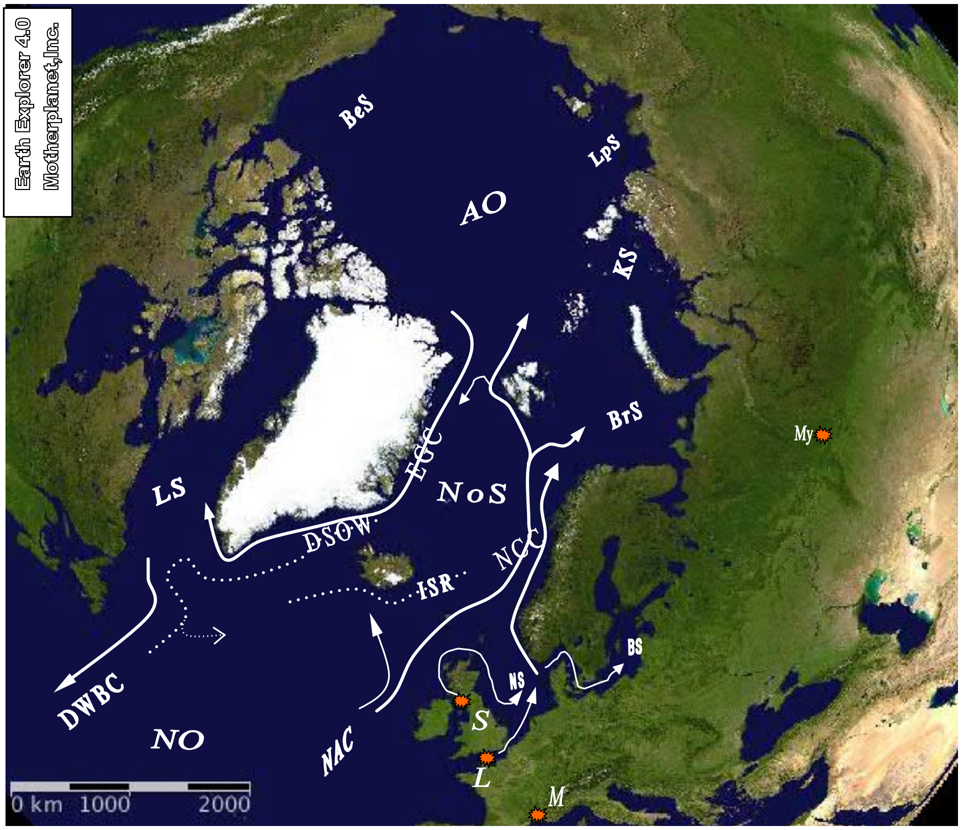

Fig. 1. Major pathways of marine discharges from Sellafield and La Hague nuclear reprocessing facilities to the Arctic and North Atlantic Oceans. Solid lines = surface currents, dashed lines = deep currents, AO = Arctic Ocean, NAO = North Atlantic Ocean, NAC= North Atlantic Current, NS = North Sea, BS = Baltic Sea, NCC = Norwegian Coastal Current, NoS = Nordic Seas, BrS = Barents Sea, KS = Kara Sea, LpS = Laptev Sea, BeS = Beaufort Sea, EGC= East Greenland Current, DSOW = Denmark Strait Overflow Water, ISR = Iceland-Scotland Ridge overflow water, LS = Labrador Sea, DWBC = Deep Western Boundary Current, S = Sellafield, L = La Hague, M = Marcoule, My = Mayak.

Mayak (Fig. 1), but presently there is no data that allows for an acceptable estimate of these contributions to the inventory in the Arctic Ocean.

3. Type of data

For the sake of uniformity, in at least one parameter (analytical technique), the authors chose to use only ^{129}I data that have been obtained by accelerator mass spectrometry from both published results and analyses made in this investigation. Samples analyzed in this investigation are presented in Table 2 and the analytical details are described below.

The extraction of I from the water and ice was performed on 100 mL samples that were filtered through a 0.45 μm membrane filter. About 1 mg of I was added as a carrier and the sample was then reduced with $NaHSO_3$ to convert IO_3^- to I^- and acidified to pH 2 with further extraction of I into $CHCl_3$, back extraction into water, and precipitation as AgI.

Extraction of I from sediments was performed on a sample of about 300 mg that was dried at room temperature in a desiccator for 12 h. The sample was put into a quartz boat and about 2.0 mg I was added as a carrier. The quartz boat was then placed in the combustion system for extraction of I. The combustion was conducted in an O_2 atmosphere at a final temperature of 800 °C for about 1 h. Iodine was trapped in a KOH–Na_2SO_3 solution and precipitated as AgI.

The AgI was mixed with Nb powder and pressed into cupper holders for the AMS measurement. Determination of ^{129}I/^{127}I in the samples was conducted at the Uppsala 5 MV Pelletron Tandem Accelerator. The reference material 4949C. Iodine-129 of the National Institute for Standards and Technology (NIST) was used as a standard.

Table 2
Samples and ^{129}I concentrations analyzed in this study

Archive	Sampling date	Depth	^{129}I (10^8atoms)
			(L^{-1})
Sea ice (Fram Strait)			
Ice core 81.96N −14.11E	May 2002		
Ice		0–20 cm	5.32 ± 0.37
Ice		60–80 cm	5.54 ± 0.35
Ice		120–140 cm	3.59 ± 0.24
Brine water in ice core (salinity 46)		100–120	22.6 ± 0.11
Sea water (at ice–water interface)		145cm	21.0 ± 0.1
Ice core 82.35N–3.09E, ice	May 2002	0–50 cm	2.48 ± 0.16
Ice core 81.44N–4.07W, ice	May 2002	0–20 cm	0.41 ± 0.08
Ice		40–60 cm	3.57 ± 0.24
Ice		190–210 cm	1.61 ± 0.18
Ice		300–320 cm	1.66 ± 0.12
Brine water in ice core (salinity 42)		300 cm	12.6 ± 0.5
Snow (Fram strait)	May 2002		4.95 ± 0.33, 3.10 ± 0.27
Storfjorden (water depth 10–266m)	May 2002		19–23
Greenland ice (72.56°N–37.61°W) (period 1900–1985)	9 samples		0.01–0.1
River water (Iceland)	June 2003 (4 samples)		0.05–0.5
Lake water			
Lakes Näsijärvi and Orivesi (Finland)	July 2004 2 samples	Surface	1–5
Lake Vättern (Sweden)	March–July 2004 (2 samples)	Surface	4–6
Lake Baikal (Russia)	August 2001 (3 samples)	Surface	0.01–0.1
Lake Biwa (Japan)	September 1999 (2 samples)	Surface	0.1–0.5
			(g^{-1})
Soil (Uppsala, Sweden)	June 2004	2 cm	3–5
Lake sediment (Loppesjön Helsingland, Sweden)	February 2003	2 cm	15
Baltic Sea sediment (59.02°N–19.03°E)	June 2002	2 cm	25

Background and memory effects were evaluated through carrier I blanks that produced a procedural background $^{129}I/^{127}I$ value $<3 \times 10^{-13}$. The instrument background was evaluated through use of a natural AgI (iodargyrite), which resulted in a value of 10^{-14}. The $^{129}I/^{127}I$ values of the samples were 50–150 times higher than the blank value thus resulting in negligible (<2%) background correction. Further details of the I chemical separation and AMS measurements are given in Alfimov et al. (2004a) and Englund et al. (in press).

Because of the relatively large variability in the published data in terms of number of samples, sampling method and period, representativeness for a reservoir and availability as ^{129}I concentration or $^{129}I/^{127}I$ values, in some cases average expected value are used without a specific reference. However, references of critical relevance to the ^{129}I concentration range in different environmental systems considered here are mentioned throughout the text.

4. Modeling and assessment

A variety of approaches are applied in calculating the amount of anthropogenic ^{129}I in the Earth's surface reservoirs for the period 1952–2004. The inventory calculation has been limited to the northern hemisphere and primarily to regional dispersal within Europe, the North Atlantic and Arctic Oceans and related Seas (Fig. 1). The main principles of the model depend on (1) the recognized global geochemical cycle of I (Kocher, 1981; Fabryka-Martin et al., 1985; Fuge, 1996) with the marine reservoir as the main (>90%) sink for I; (2) gaseous and liquid discharges from the Sellafield and La Hague nuclear reprocessing facilities are the main (>90%) source of anthropogenic ^{129}I; (3) recycling of ^{129}I between the atmosphere, lithosphere, hydrosphere and biosphere, as well as the inventory in the biosphere, is not considered; (4) the input data for inventory calculation represent both averaging and scaling of available measurements on environmental samples to the year 2004; (5) no inventory estimate of the marine atmosphere is considered, but it is expected to represent only a minor fraction (<1%) of the total inventory for the marine system and (6) global atmospheric transport of ^{129}I release from the reprocessing facilities is expected to be both limited and to represent only minute quantities of the total isotope input to the Earth's surface.

Presently there is no evidence for ^{129}I marine transport beyond the North Atlantic. Additionally, relative absence of published data on ^{129}I release from nuclear reprocessing facilities other than those listed in Table 1 makes tracing of the signal in oceans other than the Arctic and North Atlantic highly speculative. Furthermore, two assumptions were considered for estimating the marine inventory and include; (1) losses of I from marine surface water to the atmosphere are on the order of <1‰ per year (Wong, 1991; Baker, 2004), which implies a conservative behavior over the time scales considered here; (2) iodate reduction, a process that may affect I budgets at different depths, is apparently not a significant process at high latitudes (Truesdale et al., 2000).

The inventory model calculation adapted here can be divided into two main portions as land and marine reservoirs. Most of the published data represent sampling periods before 2002 and thus scaling following the models shown in Fig. 2 and actual ^{129}I release data was used to extend the inventory until 2004. The data and modeling of Hou et al. (2000) on seaweed also support this scaling approach. Areas of bare rock surfaces, mountainous regions and big lakes were excluded from the soil cover inventory. The ^{129}I in dry fallout (air and aerosols; Santos et al., 2005) has been considered to be negligible for the inventory observed in precipitation. The ^{129}I flux in precipitation over Europe represents <0.01% of the annual total releases from the reprocessing facilities (Persson et al., in press). However, model results (Fig. 2a) suggest rather insignificant changes in ^{129}I atmospheric concentration since the start of releases, which also seem to agree with the data of Buraglio et al. (2001a) and Schnabel et al. (2001) and thus the precipitation inventory was not scaled to variability in release rate function. The data of Schnabel et al. (2001) for the years 1994–1997 indicate about a 10 times higher annual flux of ^{129}I in precipitation in central Europe compared to the estimate by Buraglio et al. (2001a) for the years 1998 and 1999 in Northern Europe. In the present study, a recent estimate by Persson et al. (in press), which is based on more data sets in Europe and thus provides better the representative annual flux value for ^{129}I in precipitation over Europe, has been used.

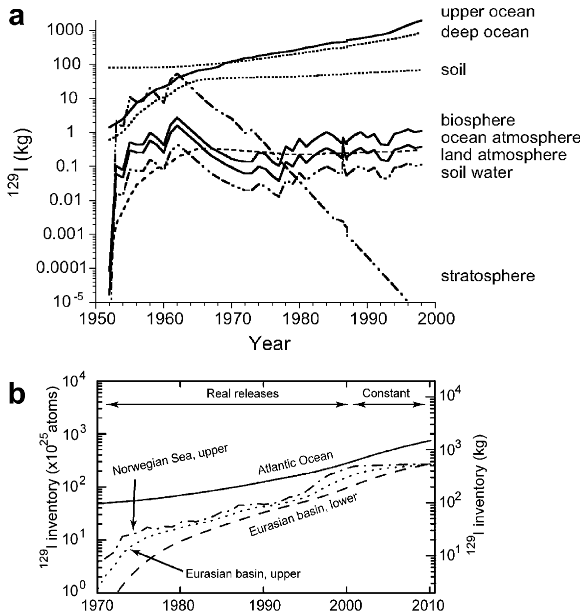

Fig. 2. (a) Modeled global anthropogenic ^{129}I inventory for the period 1952–2000 (Buraglio, 2000). Note the relatively small variability in the terrestrial compared with the marine reservoir. (b) Modeled marine distribution of anthropogenic ^{129}I in waters of the Arctic and Atlantic Oceans and the Norwegian Sea for the period 1970–2010. Note the rather small differences in the inventory between 2000 and 2010 (Alfimov et al., 2006).

Published data on ^{129}I in inland water systems (precipitation, rivers, lakes and glaciers) of Europe are scarce both spatially and temporally. The average value for rivers of the Baltic region (Aldahan et al., 2006), which is about one order of magnitude higher than the average for rivers in the USA (Moran et al., 2002), are used here. However, some exceptionally high ^{129}I concentrations (about three times higher than the value used here) have been reported from the Rhone, Rhine and Thames rivers as well as in a few small rivers in Southern Sweden (Moran et al., 2002; Snyder and Fehn, 2004; Aldahan et al., 2006).

The data on magnitude of ^{129}I in lake waters are severely limited to some from Northern Europe (Buraglio et al., 2001b), which form the basis for an average estimate used here. A more global data set is available from Snyder and Fehn (2004), but unfortunately most of the data is on lakes from continents other than Europe. Furthermore, the data of Snyder and Fehn (2004) clearly indicate about 2 orders of magnitude less concentration of ^{129}I than that expected for European lakes.

Concentration of ^{129}I in glacier ice was estimated using the available data of Wagner et al. (1996), the data on the Greenland ice cap is reported in Table 2.

There are some data on ^{129}I occurrence in the European soil cover (Hou et al., 2003; Ernst et al., 2003; Michel et al., 2005) and an average estimate, which is based on these reports, has been used. Similarly, the data on ^{129}I concentration in lake sediments seems to be presently limited to only one core from a lake in Ireland, which is relatively close to the Sellafield facility (Gallagher et al., 2005). River sediment is another reservoir that is expected to provide ^{129}I input to lakes and marine sediments. Presently, there are no data on ^{129}I concentrations of river sediment in Europe, thus the data from river waters (Aldahan et al., 2006) provided a baseline to estimate the average concentration used here.

Although distribution of ^{129}I in the marine reservoir has been more studied compared to other reservoirs, the available data have many gaps with respect to both spatial and temporal resolution. Additionally, as with the terrestrial system of Europe, data on ^{129}I concentrations in marine sediments of the region considered here are scarce. The available data include, one core from the Skagerrak sampled during 1984 (López-Gutiérrez et al., 2004), one analysis of a sediment sample from the Irish Sea (Hou et al., 2003), one core from the west North Atlantic (Moran et al., 1998) and the authors' average of two surface sediment samples from the Baltic Sea (Table 2). Consequently, the estimate of ^{129}I inventory in marine sediments is based on averaging and scaling these data, which provides a gross number. A big problem with estimation of ^{129}I in marine system relates to complicated transport pathways and time needed for the plumes to disperse and reach an intended destination. Instead, the marine inventory has been simplified into integrated accumulations that directly relate to available measured concentrations, major water current pathways (Fig. 1) and a numerical model (Fig. 2; and details in Alfimov et al., 2006) that takes into consideration transit time from the sources (Sellafield and La Hague). Thus inventories of the deep Arctic Ocean and central Nordic Seas were calculated by integrating representative ^{129}I depth profiles (Alfimov et al., 2004a,c; and unpublished data for the Norwegian Sea). For the Barents Sea inventory the estimate has been based on the data of Alfimov et al. (2004a) and Matishov et al. (2002). ^{129}I concentration of the Makarov basin deep water (Alfimov et al., 2004a) was considered for the Kara, Laptev and East Siberian Seas. In the case of the Chuckchi, Bering and Beaufort Seas, an estimate based on the data of Kilius et al. (1995) and Smith et al. (1999) was used, and data of Meili et al. (2002) and Smith et al. (2005) for the Labrador Sea. The obtained inventories were then scaled to the year 2004 by using a model-derived and time-dependent inventory (Alfimov et al., 2006). The poorly quantified ^{129}I in sea-ice cover is another challenge for the inventory in the Arctic Ocean. Here an estimate based on some measurements (Table 2), which demonstrated that sea-ice would make only a small contribution to the total inventory in the Arctic Ocean and related Seas (the Nordic, Barents and Labrador Seas) was used.

5. Results and discussion

Results of inventory calculation indicate that most (>90%) of the anthropogenic ^{129}I in the surface environmental systems is located within the marine reservoir (Table 3). The data show also a wide range of variability in ^{129}I inventory both on land and in the marine system. Soil cover contains most of the continental ^{129}I pool, whereas only minor quantities occur in the land water system. In the marine system, the Central Norwegian Sea, the Arctic Deep Water and the North Atlantic contain most of the marine inventory.

5.1. Inventory of the terrestrial system

Considering a constant ^{129}I concentration in precipitation between 1952 and 2004 and using the average annual flux, a total inventory of about 150 kg has been deposited in the terrestrial environment of Europe. This amount of deposition is comparable to estimates presently found in the terrestrial reservoirs of Europe (Table 3) and seems also to be corroborated by the relatively small ^{129}I losses by river runoff, the atmosphere and groundwater. Data from the Baltic region indicate that about 10–15% of the ^{129}I deposited by precipitation was further transported by river water to the Baltic Sea (Aldahan et al., 2006). Thus transport of ^{129}I from the terrestrial to the marine environment by runoff is relatively small even with the addition of river sediment (Table 3). Accordingly, ^{129}I losses from the terrestrial system comprise only a minor part of the residual inventory. The total inventory

Table 3
Inventory data of anthropogenic ^{129}I in the terrestrial system of Europe and in the marine system of the Arctic and North Atlantic Oceans and related Seas

Region	^{129}I (10^8 atoms)	Volume (km^3)	Surface area (km^2)	Depth (m)	Inventory atoms (kg)
Europe					
Annual precipitation	20 L^{-1}		1.05×10^7		1.3×10^{25}(3)
Rivers annual runoff	10 L^{-1}	3.1×10^3			3.1×10^{24}(0.7)
Lakes water	10 L^{-1}	1.5×10^3			1.5×10^{24}(0.3)
Glaciers (ice)	0.5 kg^{-1}		1.6×10^4	1	6.4×10^{20}(<0.01)
Surface soil	3 g^{-1}		8.91×10^6	0.1	7.0×10^{26}(150)
Lake sediment	1 g^{-1}		1.5×10^5	0.05	1.1×10^{24}(0.2)
River sediment	5 g^{-1}	680×10^6 MT			3.4×10^{23}(0.1)
Total					7.5×10^{26}(155)
Greenland (ice cape)	0.1 kg^{-1}		2.2×10^6	1	1.8×10^{22}(<0.01)
English channel					
Water	1000 L^{-1}	3.8×10^4			3.4×10^{26}(73)
Sediment	1000 g^{-1}		7.5×10^4	0.05	5.6×10^{26}(120)
Irish Sea					
Water	1000 L^{-1}	2.4×10^3			2.4×10^{26}(51)
Sediment	1000 g^{-1}		4.7×10^4	0.05	3.5×10^{26}(75)
Baltic Sea					
Water	32 L^{-1}	2.2×10^4			7.0×10^{25}(15)
Sediment	10 g^{-1}		4.2×10^5	0.05	3.2×10^{25}(7)
Skagerrak					
Water	450 L^{-1}	6×10^3			2.7×10^{26}(58)
Sediment	75 g^{-1}		6.1×10^4	0.05	3.4×10^{25} (7)
North Sea					
Water	100 L^{-1}	4×10^4			4.0×10^{26}(86)
Sediment	10 g^{-1}		5.2×10^5	0.05	3.9×10^{25} (8)
Nordic Sea					
Sediment	1 g^{-1}		2×10^6	0.05	1.5×10^{25}(3)
Water[a]					
NCC southern	114 L^{-1}		6.7×10^4	115	8.9×10^{25}(19)
NCC northern	171 L^{-1}		3.2×10^4	115	6.3×10^{25}(14)
CNS[b]	4.6 L^{-1}	4.1×10^6			1.9×10^{27}(403)
CNS (maximum)[c]	9 L^{-1}	4.1×10^6			3.7×10^{27}(792)
WSC	40 L^{-1}		1.8×10^5	500	3.6×10^{26} (77)
EGC northern	11 L^{-1}		1.7×10^5	250	4.7×10^{25}(10)
EGC central	11 L^{-1}		7×10^4	250	2.0×10^{25}(4)
EGC northern	11 L^{-1}		6.3×10^4	250	1.8×10^{25}(4)
Nordic Sea (model)[d]					5.1×10^{27}(1100)
Labrador Sea					
Water mass1	I L^{-1}		6×10^5	0–3000	1.8×10^{26}(39)
Water mass 2	3 L^{-1}		6×10^5	3000–3250	4.5×10^{25}(10)
Arctic Ocean					
Sediment	1 g^{-1}		1.5×10^7	0.05	1.1×10^{26}(24)
Ice	4 kg^{-1}		7.5×10^6	2	4.8×10^{24}(1)
Water[e]					
Southern Barnets Sea	90 L^{-1}	1.3×10^5			1.2×10^{27}(257)
Northern Barents Sea	22 L^{-1}	2.3×10^5			5.1×10^{26}(109)
Kara, Laptev and east Siberian Seas	26 L^{-1}	2.02×10^5			5.3×10^{26}(113)
Chuckchi and Beaufort Seas	0.4 L^{-1}	7.2×10^4			2.9×10^{24}(0.5)
Arctic deep water	3 L^{-1}	1.35×10^7			4.1×10^{27}(878)

Table 3 (*continued*)

Region	^{129}I (10^8 atoms)	Volume (km^3)	Surface area (km^2)	Depth (m)	Inventory atoms (kg)
North Atlantic Ocean					
Sediment	0.1 g^{-1}		4.1×10^7	0.05	3.1×10^{25}(66)
Water	0.2 L^{-1}	1.62×10^8			3.2×10^{27}(685)
Total (marine)					1.7×10^{28} (3700)

NCC = Norwegian Coastal Current CNS = Central Norwegian Sea, WSC = West Spitsbergen Current, EGC = East, Greenland Current. Precipitation 600 mm year^{-1}, soil density 2.65 g cm^{-3}, lake and marine sediment density 1.5 g cm^{-3}, density of firn 0.66 g cm^{-3}, density of ice 0.80 g cm^{-3}. Lake surface area is considered as 1.5% of Europe with an average depth of 10 m (details available in Kristen and Hansen, 1994). Estimate of river runoff is from Global Runoff Data Center, Federal Institute of Hydrology, Germany (Dr. Thomas Maurer) and from Dai and Trenberth (2002). Estimate of river sediment discharge (million tons) is from Syvitski et al. (2005). MT = metric tons.

[a] Inventory values for the NCC and WSC waters refer to their surface area multiplied by average ^{129}I concentrations in depth profiles measured by Alfimov et al. (2004a,c) and unpublished data.

[b] The inventory of the CNS was estimated from a linearly-interpolated ^{129}I profile sampled in 2002 in the center of the Greenland Sea (Alfimov et al., 2004c) multiplied with an area of CNS and scaled to 2004 with help of the modeled inventories (see Fig. 2b) by Alfimov et al. (2006). Hence, the "^{129}I concentration of CNS" is an effective average concentration derived from the estimated inventory and the volume of CNS and is therefore only for a mere curiosity of the reader.

[c] For comparison, we give an estimated upper limit for the ^{129}I inventory of the CNS derived from just ^{129}I surface concentration and the volume of the CNS.

[d] For comparison, we give ^{129}I inventory in 2004 modeled for the whole region of the Nordic Seas (+NCC, WSC, EGC) by Alfimov et al. (2006).

[e] Estimated from the depth profiles by Alfimov et al. (2004a).

of ^{129}I in the terrestrial system (sediment, water and ice) represents about 3% of the total releases from the European reprocessing facilities and about 50% of the gaseous releases (Tables 1 and 3). Apparently, the amount of ^{129}I from gaseous release can account for the inventory in the terrestrial system of Europe, but the question is whether this is the most common source and transport mode of the isotope. Unlike the weapon tests and to a certain extent the Chernobyl accident, the injection of ^{129}I into the atmosphere from the reprocessing facilities is not explosive and does not reach high altitudes. It may reach up to a few hundred meters and thus its spread is strongly dependent on wind strength, direction and moisture content. Accordingly, it is expected that a large part of ^{129}I from gaseous release would be deposited in areas close to reprocessing facilities as also indicated by a few studies (Rucklidge et al., 1994; Fréchou and Calmet, 2003). However, even if the exponential decrease of soil iodine is considered (Ernst et al., 2003) and the European soil inventory is recalculated to a depth of 25 cm instead of the 10 cm considered here, about 30% of the gaseous release will still be missing.

Another source of ^{129}I to the atmosphere is releases from oceans and seas as sea spray, organic and inorganic forms (Wong, 1991; Baker, 2004). Data concerning environmental ^{127}I and ^{129}I distribution show depletion of I in inland areas away from coastal regions in both precipitation and runoff (Moran et al., 1999; Szidat et al., 2000; Persson et al., in press). This behavior suggests that a portion of atmospheric ^{129}I in the atmosphere of Europe can be related to marine sources, but it is difficult to quantify the contribution of each source (marine or direct gaseous releases from the reprocessing facilities) to the total terrestrial inventory.

The amount of ^{129}I stored in glaciers and the Greenland ice cap is minute due to the low concentration (Table 3). However, it is still expected that the isotope concentration will be highly variable depending on the location of the glacier with respect to latitude, altitude and proximity to coastal regions. These factors affect ^{129}I concentration in precipitation and thus it is estimated that more ^{129}I will be accumulated in the glaciers of Scandinavia than those in the Alps. Furthermore, survey data (Wagner et al., 1996 and Table 2 of this study) show relatively elevated concentrations in the top parts of glaciers and the Greenland ice that can act as ultra sensitive indicators of the anthropogenic era contribution and eventually provide a new tool for understanding glacier dynamics and climate change.

Unlike the water system, regional and local geochemical variability of the soil cover in Europe may strongly affect the estimated soil inventory (Ernst et al., 2003). Generally, acid soils may lose more iodine to the atmosphere and ground water than alkaline soils (Fuge, 1996). However, available data on ^{129}I in soil show that most of the isotope concentration is retained within the upper 5–10 cm (Ernst et al., 2003; Michel et al., 2005) while there is some loss to ground water. The inventory of the isotope in lake sediments, although of small magnitude (<0.1% of the terrestrial system), must be considered with caution. Nevertheless, the isotope distribution in lake sediment can provide a new time marker for atomic-era emissions since the forties and consequently facilitates estimation of sedimentation rates. The inventory of river sediments can vary due to the expected depth profile that can be a few centimeter to a few meters due to the strongly dynamic sedimentation and erosion processes in rivers and catchment areas. Therefore, data on ^{129}I concentrations in river sediments are urgently needed not only for recognition of the isotope inventory, but also they can provide interesting information about recent (last 60 a) soil erosion in drainage basins.

5.2. Inventory of the marine system

As about 90% of the ^{129}I inventory occurs in the marine reservoir, which, together with relatively better data sets and a more homogenous environment than the terrestrial system, forms a more solid base for discussing the isotope distribution patterns. Apart from the generally decreasing concentration of I with depth in marine waters, there is large variability, by several orders of magnitude, even within the surface and deep waters. The highest inventory of the isotope is still, however, within the surface layers of all the water bodies considered in Table 3, except in the Baltic and the Labrador Seas where the deep layer is more enriched than the surface layer (Alfimov et al., 2004b; Smith et al., 2005). Most of the ^{129}I inventory seems to reside in the Nordic Seas and the Arctic Ocean that contain together about 70% of the total inventory. Investigations of these water bodies also suggest both continuous increase and transport of ^{129}I to the North Atlantic via mainly the Labrador Sea and Denmark Strait (Meili et al., 2002; Alfimov et al., 2004b; Smith et al., 2005). Presently there is relatively more systematic data about the temporal distribution of ^{129}I in the Arctic Ocean, Nordic, Labrador and Baltic Seas than in the North and Irish Seas. Accordingly, the inventory estimation for the latter seas are expected to be subject to change if new data are acquired. It is likely that the part of the basins in the vicinity of the facilities will contain relatively larger quantities of the isotope than estimated here.

The disequilibrium between ^{129}I and ^{127}I, and the relatively long residence time (10 ka) of I in the oceans strongly affects the dynamics of inventory within the ocean water layers. Although temporal variability in ^{129}I concentration between the different ocean water layers may exist, it is expected that the total inventory of a whole water column will be somewhat representative, at least in response to transient time of water currents as discussed here. Based on the general water circulation from Fig. 1, the plume from the Sellafield marine discharge enters the North Atlantic west of Scotland and is swept into the North Sea by the NAC. ^{129}I data have apparently identified the Sellafield plume NW of the UK admixed into the coastal branch of the NAC, before it enters the North Sea (Alfimov et al., 2004b). This is evident from the salinity profile, which maintains values of the NAC whereas the ^{129}I value sharply increases (20 times). This feature gives a specific character for this surface water parcel of the NAC which otherwise seems homogenous in terms of salinity. The ^{129}I-labelled water diverts into the North Sea, whereas most of the NAC continues into the Norwegian Sea. The other marine source of ^{129}I to the North Sea is the discharge from the La Hague facility entering through the English Channel. Surface circulation in the North Sea is highly variable locally, but a major path of the ^{129}I outflow is along the coast of Denmark and into the Norwegian Coastal Current (e.g. Dahlgaard et al., 1995; Alfimov et al., 2004b). The inventory estimate for the North Sea is rather poor because of scarce data points and the about 100 kg of ^{129}I (Table 3) may be dominated by the La Hague releases. However, at this stage of the investigation, and without a more detailed data set from the western and eastern coasts of the North Sea, it is quantitatively difficult to separate the Sellafield and La Hague contributions to the outflow current and eventually to changes in the inventory of ^{129}I. For simplicity, a single surface water outflow (discharge parcel) from the North Sea has been considered for the releases from the two facilities and estimation of transient times and inventory in the Nordic and Labrador Seas and the Arctic and North Atlantic Oceans.

The expected flux and transit time for each compartment of water masses reaching the North Atlantic Ocean from the Nordic and Labrador Seas, and the Arctic Ocean were considered in detail by

Smith et al. (2005) and Alfimov et al. (2006). The inflow of ^{129}I to the Arctic Ocean is by the ^{129}I-enriched surface branch of the NAC and the North Sea water mass along the Norwegian coast (Gascard et al., 2004). Inventory estimates for the Norwegian Coastal Current (NCC) sum up to about 35 kg of ^{129}I (Table 3), which is about 30–40% the amount found in the North Sea. Estimates of inventory for the Nordic Seas water can vary between 530 and 920 kg, which is about 20–50% less than the estimate made by Alfimov et al. (2006) based on numerical modeling. The 20% difference can be explained by error estimates, but obviously more data on ^{129}I distribution in the CNS are needed for constrained comparison between the model and empirical approach.

Transit time of ^{129}I from the source (considered here as the "feeder" currents from Sellafield and La Hague) to the Arctic Ocean through the Norwegian Sea is estimated at about 6–11 a (Buraglio et al., 1999; Hou et al., 2000; Edmonds et al., 2001; Smith et al., 2005). However, the estimated inventory for the different parts of the Arctic Ocean water is about 1300 kg of ^{129}I, which is about 30% more than in the Norwegian Sea and represents about 30% of the total liquid releases from the nuclear reprocessing facilities (Table 1) until the year 2004. The estimates for the Arctic Ocean ^{129}I inventory presented here seem, however, comparable to the data produced by numerical modeling (Alfimov et al., 2006).

Transport of ^{129}I from the Arctic Ocean occurs mainly by cold, relatively fresh, waters that enter the Greenland Sea (East Greenland Current, EGC) through the Fram Strait. This water mass overflows the Denmark Strait (Denmark Strait Overflow Water; DSOW) and together with the cold water from the Norwegian and Iceland Seas forms part of the Deep Western Boundary Current (DWBC) (Pickart, 1992; Dickson and Brown, 1994; Smethie and Fine, 2001). The DWBC comprises the return mode of the "conveyer belt" in the North Atlantic. Inventory data presented in Table 3 indicate occurrence of about only 20 kg in the EGC water, which is about one order of magnitude less than that predicted by the numerical modeling of Alfimov et al. (2006). A possible reason for the difference is strong influence from the Central Norwegian Sea (CNS) and returned WSC that may rapidly convey high amounts of ^{129}I through the EGC to the Labrador Sea and the DWBC after a relatively short residence time. Accordingly, the inventory of EGC presented in this study is most likely underestimated. This can also be tested through comparison with the ^{129}I pool in the Labrador Sea before final incorporation into the DWBC and the western North Atlantic. A transit time of 1 to a few years (e.g. Smethie and Fine, 2001; Smith et al., 2005) is attributed to transport of ^{129}I from the Arctic Ocean to the Labrador Sea, which means more delay in the inventory signal. However, the results in Table 3 suggest about 40 kg of ^{129}I in the Labrador Sea; about double the amounts in the EGC.

The DWBC may represent the main pathway of waters enriched with ^{129}I that enter the West North Atlantic and may form a unique tracer of further water mass transport along the North American coast and further into the North Atlantic Gyre, and the subtropical–tropical exchange water masses. Data from the Labrador Sea deep water (Edmonds et al., 2001; Smith et al., 2005), the Mid Atlantic Bight (Santschi et al., 1996) and the Gulf of Mexico (Schink et al., 1995) indicate elevated ^{129}I concentration in these water masses and are thus in agreement with the proposed trend. Furthermore, there are no data on ^{129}I concentration in the return cold water from the Norwegian Sea (flow over the Iceland–Scotland Ridge, ISR), but it has been assumed that these water masses contain an order of magnitude lower concentration than the DSOW. A flux of ^{129}I has been estimated, which is based on mean annual transport rates of about 8 Sv, 4 Sv and 2 Sv ($1\ \mathrm{Sv} = 10^6\ \mathrm{m^3/s}$) for the NAC-NS, DSOW at south Greenland and ISR, respectively (Smethie and Fine, 2001; Orvik et al., 2001). The authors have used ^{129}I values of 2×10^9 atoms/L for the NAC-NS (average of NAC-NS measured values; Alfimov et al., 2004a) and 1×10^9 atoms/L for the DSOW (Averaged measured values; Alfimov et al., 2004c) and 0.5×10^8 for ISR (assuming values similar to the 1000–2000 m deep water of the Greenland and Norwegian Seas; Alfimov et al., 2004c; Gascard et al., 2004; Smith et al., 2005), although the ISR contribution to the total inventory should be considered with caution. Average annual flux of ^{129}I, at about 100 kg from the NAC-NS, 26 kg for the DSOW and 0.5 kg for ISR since 1999, can be estimated. Using this gross approximation, about 10–20% of the total ^{129}I entering waters of the Norwegian Sea may be currently back-ventilated into the DWBC. The remainder of the ^{129}I emission is most likely residing in the Arctic Ocean and Nordic Seas.

10–20% of the total inventory in the Nordic Seas represents about 100–200 kg ^{129}I, which is only 10–20% of the total inventory for the North Atlantic (Table 3). The inventory estimate for the North Atlantic Ocean presented in Table 3 seems, however, to be about 30% lower than the one estimated from numerical modeling (Alfimov et al., 2006).

6. Conclusions

The total estimated ^{129}I inventory for all compartments or regions considered in Table 3 suggests some (about 1000 kg) missing amount compared to the sum of the total releases from Sellafield and La Hague facilities (Table 1). Most of this difference between inventory and release data seems to occur in the marine system. The plausible cause of this difference may relate to underestimated inventories in the marine area, particularly the sediment compartment, in close proximity to the reprocessing facilities, such as the Irish Sea, English Channel and the North Sea. However, it is important to remember that part of the isotope inventory occurs within the biosphere, which has not been considered here. Uncertainty in release data from the reprocessing facilities may also contribute to the difference between the inventory and total releases. Each of these scenarios seems interesting and further research is urgently needed in order to better understand and utilize ^{129}I as a tracer of environmental processes. The long half-life (15.7 Ma) of the isotope adds a significant advantage in understanding ocean circulation and eventual impact on future climate change.

Acknowledgements

The authors thank P. Santschi, S. Szidat and an anonymous reviewer for constructive review comments.

References

Aldahan, A., Kekli, A., Possnert, G., 2006. Distribution and sources of ^{129}I in rivers of the Baltic Region. J. Environ. Radioact. 88, 49–73.

Alfimov, V., Aldahan, A., Possnert, G., Windsor, P., 2004a. Anthropogenic iodine- ^{129}I in seawater along a transect from the Norwegian coastal current to the North Pole. Mar. Poll. Bull. 49, 1097/1104.

Alfimov, V., Aldahan, A., Possnert, G., Kekli, A., Meili, M., 2004b. Concentration of ^{129}I along a transect from the North Atlantic to the Baltic Sea. Nucl. Inst. Meth. Phys. Res. B223–224, 446–450.

Alfimov, V., Aldahan, A., Possnert, G., 2004c. Tracing water masses with ^{129}I in the western Nordic Seas in early spring 2002. Geophys. Res. Lett. 31, L19305.

Alfimov, V., Aldahan, A., Possnert, G., 2006. Anthropogenic iodine-129 in the Arctic Ocean and Nordic Seas: numerical modeling and prognoses. Mar. Poll. Bull. 52, 380–385.

Baker, A.R., 2004. Inorganic iodine speciation in tropical Atlantic aerosol. Geophys. Res. Lett. 31, L23S02.

BNFL, 1999–2004. British Nuclear Fuels, Annual Discharge Reports. Available from: http://www.bnfl.co.ukhttp://www.bnfl.co.uk.

Buraglio, N., 2000, Accelerator mass spectrometry of ^{129}I and its applications in natural water systems. Ph.D. thesis, Uppsala University.

Buraglio, N., Aldahan, A., Possnert, G., 1999. Distribution and inventory of ^{129}I in the central Arctic Ocean. Geophys. Res. Lett. 26, 1011–1014.

Buraglio, N., Aldahan, A., Possnert, G., Vintersved, I., 2001a. ^{129}I from the nuclear reprocessing facilities traced in precipitation and runoff in northern Europe. Envir. Sci. Technol. 35, 1579–1586.

Buraglio, N., Aldahan, A., Possnert, G., 2001b. ^{129}I in lakes of the Chernobyl fallout region and its environmental implications. Appl. Radiat. Isotopes 55, 715–720.

Cogema, 1997. Rapport Environment, Cogema Marcoule, 1997.

Dahlgaard, H., Chen, Q., Herrmann, J., Ibbett, R., 1995. On the background level of ^{99}Tc, ^{90}Sr and ^{137}Cs in the North Atlantic. J. Mar. Syst. 6, 571–578.

Dai, A., Trenberth, K., 2002. Estimate of freshwater discharge from continents: latitudinal and seasonal variations. J. Hydrometeorol. 3, 660–687.

Dickson, R.R., Brown, J., 1994. The production of North Atlantic Deep Water Sources, rates and pathways. J. Geophys. Res. 99, 12319–12341.

Edmonds, H.N., Zhou, Z.Q., Raisbeck, G.M., Yiou, F., Kilius, L.R., Edmonds, J.M., 2001. Distribution and behavior of anthropogenic ^{129}I in water masses ventilating the North Atlantic Ocean. J. Geophys. Res. 106, 6881–6894.

Eisenbud, M., Gesell, T., 1997. Environmental Radioactivity. 4th ed.. Academic Press, San Diego.

Englund, E., Aldahan, A. Possnert, G., Alfimov, V. in press. A routine preparation method for AMS measurement of ^{129}I in solid material. Nucl. Inst. Meth. Phys. Res. B.

Ernst, T., Szidat, S., Handl, J., Jakob, D., Michel, R., Schnabel, C., Synal, H.-A., Santos, F.J., Benne, I., Boess, J., Gehrt, E., Capelle, A., Schneider, J., Schäfer, W., Böttcher, J., 2003. Migration of iodine-129 and iodine 127 in soils. Kernteknik 68 (4), 155–167.

Fabryka-Martin, Bently H., Elmore, D., Airey, P., 1985. Natural iodine-129 as an environmental tracer. Geochim. Cosmochim. Acta 49, 337–347.

Fréchou, C., Calmet, D., 2003. ^{129}I in the environment of the La Hague nuclear fuel reprocessing plant – from sea to land. J. Environ. Radioact. 70, 43–59.

Fuge, R., 1996. Geochemistry of iodine in relation to iodine deficiency diseases. In: Appleton, J.D., Fuge, R., McCall, J.H. (Eds.), Environmental Geochemistry and Health, vol. 113. Geological Society Special Publication, pp. 201–211.

Gallagher, D., McGee, E., Mitchell, P., Alfimov, V., Aldahan, A., Possnert, G., 2005. Retrospective search for evidence of the 1957 Windscale fire in NE Ireland using ^{129}I and other long-lived nuclides. Environ. Sci. Technol. 39, 2927–2935.

Gascard, J.-C., Raisbeck, G., Sequeira, S., Yiou, F., Mork, K.A., 2004. The Norwegian Atlantic Current in the Lofoten basin inferred from hydrological and tracer data (^{129}I) and its interaction with the Norwegian Coastal Current. Geophys. Res. Lett., L01308.

Gray, J., Jones, S.R., Smith, A.D., 1995. Discharges to the environment from the Sellafield site, 1951–1992. J. Radiolog. Protect. 15, 99–131.

GRNC, 1999. Groupe Radioécologie Nord-Cotentin: Inventaire des rejets radioactifsdes installations nucléaires: Historique des rejets, Rapport Finale Détaillé, vol. I. Groupe Radioécologie Nord-Cotentin GT1, c/o Institut de Protection et de Sureté Nucléaire, Fonteney-aux-Roses; 1999 (in French). Available from: http://www.irsn.org/Nordcotentin/francais/GRNC/Gt1/sommaire.htmhttp://www.irsn.org/Nord-cotentin/francais/GRNC/Gt1/sommaire.htm.

Hanford, 1997. The Release of Radioactive Materials from Hanford: 1944–1972. The Hanford Health Information Network website. Available from: http://www.doh.wa.gov/hanford/publications/history/release.htmlhttp://www.doh.wa.gov/hanford/.

Hou, X.L., Dahlgaard, H., Nielsen, S.P., 2000. Iodine-129 time series in Danish, Norwegian and Northwest Greenland Coast and the Baltic Sea by seaweed. Estuar. Coast. Shelf Sci. 51, 571–584.

Hou, X.L., Fogh, C.L., Kucera, J., Andersson, K.G., Dahlgaard, H., Nielsen, S.P., 2003. Iodine-129 and Caesium-137 in Chernobyl contaminated soil and their chemical fractionation. Sci. Total Environ. 308, 97–109.

Jackson, D., Lambers, B., Gray, J., 2000. Radiation doses to members of the public near to Sellafield, Cumbria, from liquid discharges 1952-98, J. Radiolog. Protect. 20, 139–167.

Kilius, L.R., Zhao, X.-L., Smith, J.N., Ellis, K.M., 1995. The measurements of ^{129}I in the Canadian Arctic Basin and other Arctic waters. In: Strand, P., Cooke, A. (Eds.), Environmental Radioactivity in the Arctic, pp. 117–120.

Kocher, D., 1981. A dynamic model of the global iodine cycle and estimation of dose to the world population from releases of ^{129}I to the environment. Environ. Int. 5, 15–31.

Kristen, P., Hansen, H., 1994. European Rivers and Lakes: Assessment of their environmental state. European Environmental Agency, monographs 1.

López-Gutiérrez, J.M., García-León, M., Schnabel, Ch., Suter, M., Synal, H.A., Szidat, S., Garcìa-Tenorio, R., 2004. Relative influence of ^{129}I sources in sediment core from the Kattegat area. Sci. Total Environ. 323, 195–210.

Matishov, G.G., Matishov, D.G., Namjatov, A.E., Smith, J.N., Carroll, J., Dahle, S., 2002. Radioactivity near the Sunken Submarine "Kursk" in the Southern Barents Sea. Environ. Sci. Technol. 36, 1919–1922. doi:10.1021/es011248.

Meili, M., Aldahan, A., Hou, X., Oughton, D.H., Possnert, G., 2002. Radioactive iodine (^{129}I) in the North Atlantic region: regional and Global atmosphere–land–sea interactions. In: The Second Nordic Arctic Research Programme (NARP) Symposium, Akureyri, Iceland, 23–24 May 2002.

Michel, R., Handl, J., Ernst, T., Botsch, W., Szidat, S., Schmidt, A., Jakob, D., Belta, D., Romantschuk, L.D., Synal, H.-A., Schnabel, Ch., Lopez- Gutierrez, J.M., 2005. Iodine-129 in soils from northern Ukrania and the retrospective dosimetry of the iodine-131 exposure after the Chernobyl accident. Sci. Total Environ. 340, 35–55.

Mironov, V., Kudrjashov, V., Yiou, F., Raisbeck, G.M., 2002. Use of ^{129}I and ^{137}Cs in soils for the estimation of ^{131}I deposition in Belarus as a result of the Chernobyl accident. J. Environ. Radioact. 59, 293–307.

Moran, J.E., Fehn, U., Teng, R.T., 1998. Variations in ^{129}I/^{127}I ratio in recent marine sediment: evidence for a fossil organic component. Chem. Geol. 152, 193–203.

Moran, J.E., Oktay, S., Santschi, P.H., Schink, D.R., 1999. Atmospheric dispersal of ^{129}I from nuclear fuel reprocessing facilities. Environ. Sci. Technol. 33, 2536–2542.

Moran, J.E., Oktay, S.D., Santschi, P.H., 2002. Sources of Iodine and Iodine-129 in Rivers. Water. Resour. Res. 38 (8), pp. 24-1 to 24-10.

Orvik, K.A., Skagseth, O., Mork, M., 2001. Atlantic inflow to the Nordic Seas: current structure and volume fluxes from moored current meters, VM-ADCP and SeaSoar-CTD observations, 1995–1999. Deep-Sea Res. 48, 937–957.

Persson, S., Alfimov, V., Aldahan, A., Possnert, G., Hau, X., in press. ^{129}I variability in precipitation over Europe. Nucl. Inst. Meth. Phys. Res. B.

Pickart, R.S., 1992. Water mass components of the North Atlantic Deep Western Boundary Current. Deep-Sea Res. 39, 1553–1572.

Raisbeck, G.M., Yiou, F., 1999. ^{129}I in the oceans: origins and applications. Sci. Total Environ. 237–238, 31–41.

Rucklidge, J., Kilius, L., Fuge, R., 1994. ^{129}I in moss down-wind from Sellafield nuclear fuel reprocessing plant. Nucl. Inst. Meth. Phys. Res. B 92, 417–420.

Santos, F.J., López-Gutiérrez, J.M., García-León, Sutter M., Synal, -H.A., 2005. Determination of ^{129}I/^{127}I in aerosol samples in Seville (Spain). J. Environ. Radioact. 84, 103–109.

Santschi, P.H., Schink, D.R., Corapcioglu, O., Oktay-Marshall, S., Fehn, U., Sharma, P., 1996. Evidence for elevated levels of iodine-129 in the Deep Western Boundary Current in the Middle Atlantic Bight. Deep-Sea Res. 43, 259–265.

Schink, D.R., Santschi, P.H., Corapcioglu, O., Fehn, U., 1995. ^{129}I in Gulf of Mexico waters. Earth Planet. Sci. Lett. 135, 131–138.

Schnabel, C., Lopez-Gutierrez, J.M., Szidat, S., Sprenger, M., Wernli, H., Beer, J., Synal, H-A., 2001. On the origin of ^{129}I in rain water near Zürich. Radiochim. Acta 89, 815–822.

Smethie, W.M., Fine, R.A., 2001. Rates of North Atlantic Deep Water formation calculated from chlorofluorocarbon inventories. Deep-Sea Res. 48, 189–215.

Smith, J.N., Ellis, K.M., Boyd, T., 1999. Circulation features in the central Arctic Ocean revealed by nuclear fuel reprocessing tracers from Scientific Ice Expeditions 1995 and 1996. J. Geophys. Res. 104, 29663–29677.

Smith, J.N., Jones, E.P., Moran, S.B., Smethie, W.M., Kieser, W.E., 2005. Iodine 129/CFC11 transit times for the Denmark Strait overflow water in the Labrador and Irminger Seas. J. Geophys. Res. 110, C05006.

Snyder, G., Fehn, U., 2004. Global distribution of ^{129}I in rivers and lakes: implications for iodine cycling in surface reservoirs. Nucl. Inst. Meth. Phys. Res. B (223/224), 579–586.

Syvitski, J., Vörösmarty, C., Kettner, A., Green, P., 2005. Impact of Humans on the flux of terrestrial sediment to the global coastal ocean. Science 308, 376–380.

Szidat, S., Schmidt, A., Handl, J., Jakob, D., Botsch, W., Michel, R., Synal, H.A., Schnabel, C., Sutter, M., López-Gutiérrez, J.M., Städe, W., 2000. Iodine-129: sample preparation, quality control and analyses of pre-nuclear materials and of natural waters from lower Saxony, Germany. Nucl. Inst. Meth. Phys. Res. B172, 699–710.

Truesdale, V.W., Bale, A.J., Woodward, E.M., 2000. The meridional distribution of dissolved iodine in near surface waters of the Atlantic Ocean. Prog. Oceanog. 45, 387–400.

UNSCEAR, 2000. Sources and Effects of Ionizing Radiation, Volume I, Annex A: Dose assessment methodologies, United Nations Scientific Committee on the Effects of Atomic Radiation.

Wagner, M.J.M., Dittrich-Hannen, B., Synal, H-A., Suter, M., Schotterer, U., 1996. Increase of ^{129}I in the environment. Nucl. Inst. Meth. Phys. Res. B 113, 490–494.

Wong, G.T.F., 1991. The marine geochemistry of iodine. Rev. Aquat. Sci. 4, 45–73.