

$^{129}\text{I}/^{127}\text{I}$ ratios in surface waters of the English Lake District

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Abstract

Accelerator Mass Spectrometry (AMS) was used to measure $^{129}\text{I}/^{127}\text{I}$ ratios in surface sea, lake, and river water samples collected in 2004 and 2005 from the English Lake District and from SW Scotland, areas which are in relatively close proximity to the Sellafield nuclear fuel reprocessing plant in NW England. The $^{129}\text{I}/^{127}\text{I}$ ratios in surface water collected from the shore of the Irish Sea were in the range 2.8×10^{-6} to 8.2×10^{-6} . These ratios are one order of magnitude higher than that of seawater collected from the Irish Sea in 1992, correlating with the increase in ^{129}I content of the Sellafield liquid effluent discharge over the last decade. The $^{129}\text{I}/^{127}\text{I}$ ratios in lakes in the Lake District were in the range 0.7×10^{-6} to 6.4×10^{-6} and decreased exponentially as a function of distance from Sellafield. Consideration of the relative variation of stable I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios suggests that Sellafield gaseous discharges may be the dominant source of ^{129}I to the lakes.

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1. Introduction

Iodine is a biophilic element which has only one stable isotope (^{127}I) and one long-lived radioisotope, ^{129}I ($t_{1/2} = 15.7 \times 10^6$ a). Naturally occurring ^{129}I is mainly produced by spontaneous fission of U in the lithosphere and by the interaction of cosmic ray particles with Xe in the upper atmosphere. The pre-nuclear era equilibrium $^{129}\text{I}/^{127}\text{I}$ ratio in the marine hydrosphere was estimated theoretically to be 5.5×10^{-13} (Fabryka-Martin et al., 1985)

while measurements of pre-nuclear era marine sediments and deep ocean waters have indicated a $^{129}\text{I}/^{127}\text{I}$ ratio of $\sim 1.5 \times 10^{-12}$ (Moran et al., 1998; Fehn et al., 1986; Schink et al., 1995). ^{129}I is also produced anthropogenically and is formed in high yield by nuclear fission of ^{235}U and ^{239}Pu . Thus, the concentrations of ^{129}I in surface reservoirs have increased as a consequence of the civil and military use of nuclear fission. The total release of ^{129}I to the atmosphere from atmospheric nuclear weapons testing since the 1940s and particularly in the early 1960s was estimated to have been 50–150 kg (UNSCEAR, 1982; Raisbeck et al., 1995; Chamberlain, 1991; Eisenbud and Gesell, 1997). After the introduction of the partial atmospheric nuclear test

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ban treaty in 1963, the main anthropogenic source of ^{129}I has been peaceful uses of nuclear energy, via releases into the environment from nuclear fuel reprocessing facilities. Because of its long half-life, ^{129}I accumulates in the environment. It is therefore essential that the distribution and movement of ^{129}I in the environment should be studied because it is potentially a radiological hazard to humans, due to its affinity for the thyroid gland. ^{129}I is also useful as a dating and tracing tool in hydro-geologic and oceanographic studies.

The main sources of ^{129}I to the environment in Western Europe are the nuclear fuel reprocessing plants at Sellafield, NW England, and La Hague, NW France. This is illustrated by ^{129}I concentrations in an ice core, collected from the Swiss Alps in 1988, which showed a continuous increase in response to releases from nuclear fuel reprocessing plants which was even larger than that for fallout from nuclear weapons tests in the 1960s (Wagner et al., 1996). Fig. 1 shows the ^{129}I content of liquid and gaseous waste emissions from Sellafield from the commencement of the operation in 1952 to 2004, and Table 1 shows the data in 1999 to 2004. The amount of ^{129}I discharged into the Irish Sea has increased steeply over the last decade. On the other hand, the amount of ^{129}I in the gaseous discharge has not varied significantly. The total discharge of ^{129}I from Sellafield in 2004 was 102 kg, and the contribution of gaseous discharge to the total was 2.4%. Liquid radioactive waste from the Sellafield nuclear fuel reprocessing plant is released from a pipeline extending 2.5 km into the Irish Sea from the high water mark. ^{129}I discharged into the

Table 1

Liquid and gaseous ^{129}I emission from Sellafield in 1999–2004 (kg $^{129}\text{I}/\text{a}$)

Year	Liquid	Gaseous
1999	73.44	3.83
2000	71.91	3.83
2001	96.39	3.06
2002	111.69	3.21
2003	84.15	2.60
2004	99.47	2.42

1999–2003: from Discharges and Monitoring of the Environment in the UK, BNFL Annual Report, 2003. 2004: from Radioactivity in Food and the Environmental Report 2004 (RIFE-10), 2005.

sea is mixed with naturally occurring stable I, and thereafter follows the biogeochemical cycles and environmental transfer pathways of stable I, with some being released from the sea to the atmosphere and transported onshore.

Several studies have used archived environmental samples to trace ^{129}I from the nuclear fuel reprocessing plants. For example, Raisbeck et al. (Raisbeck et al., 1995; Yiou et al., 1994) measured ^{129}I in samples of seaweed and seawater taken in the 1980s and early 1990s at various distances along the coasts from the nuclear fuel reprocessing facilities, including the Irish Sea, observing $^{129}\text{I}/^{127}\text{I}$ ratios in the range 5.1×10^{-7} to 8.9×10^{-7} in 1992. Schnabel et al. (2005, 2007) reported that the $^{129}\text{I}/^{127}\text{I}$ ratios in Scottish seawater from 2003 to 2005 were 7.1×10^{-8} – 3.4×10^{-6} . For freshwater in England, the $^{129}\text{I}/^{127}\text{I}$ ratios in the River Granta in Cambridge and the Thames in London were measured to be 9.8×10^{-9} and 1.9×10^{-8} in 1999 (Snyder and Fehn, 2004). Rucklidge et al. (1994) measured $^{129}\text{I}/^{127}\text{I}$ ratios in mosses collected along a 60 km transect of the English Lake District, and the results showed a remarkable variation in ratio over relatively small distances. This is for topographical and geological reasons such as shielding by hills and the influence of limestone bedrock. The $^{129}\text{I}/^{127}\text{I}$ ratio in lake water is expected to show the representative value of the whole catchment area. However, the ratio for lakes in the Lake District has not been reported.

In this study, $^{129}\text{I}/^{127}\text{I}$ ratios were measured in surface sea, lake and river water samples from the area near Sellafield, including the English Lake District and southern Scotland. The main aim of this study was to provide data for ^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios in freshwater samples near one of the world's main sources of ^{129}I . The relative

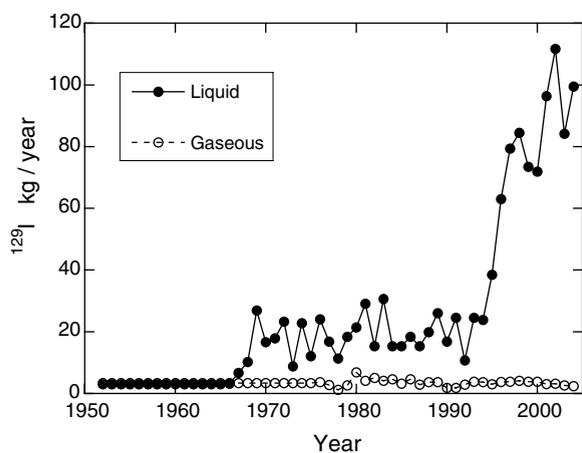


Fig. 1. Liquid and gaseous ^{129}I emission from Sellafield. Data in 1952–1998 are from López-Gutiérrez et al. (2004).

contribution of liquid and gaseous ^{129}I discharges from Sellafield to the total ^{129}I deposition in the area are also estimated. Moreover, the new ^{129}I concentrations for the Irish Sea can be used as input data for the modeling of the sea to land transfer of ^{129}I and thus for dose calculations based on that pathway.

2. Methods

2.1. Sampling

Surface water samples, of ~ 1 L, were collected in March, November and December of 2004, and May and June of 2005 from the following locations: (1) 7 lakes and 4 rivers near Sellafield in the Lake District, (2) a lake (Loch Ken) in SW Scotland where ^{129}I deposition directly from Sellafield gaseous dis-

charges is expected to be small, and (3) 4 points on the shoreline of the Irish Sea. Fig. 2 shows the locations of the sampling points and Table 2 shows the distance from Sellafield and the sea to the middle point of the lakes.

2.2. Analysis of ^{129}I

The collected water samples were made alkaline by addition of NaOH to avoid evaporative losses of I, and filtered using a $0.45\ \mu\text{m}$ membrane filter. Thereafter, the samples were split into separate sub-samples for ^{129}I and ^{127}I analysis, with about 1 L samples being used for analysis of ^{129}I in lake and river water and 40 mL samples for seawater. To dilute the ^{129}I concentration for AMS analysis, 5–15 mg of Woodward iodine carrier were added to the sample. Then, to convert all I species including

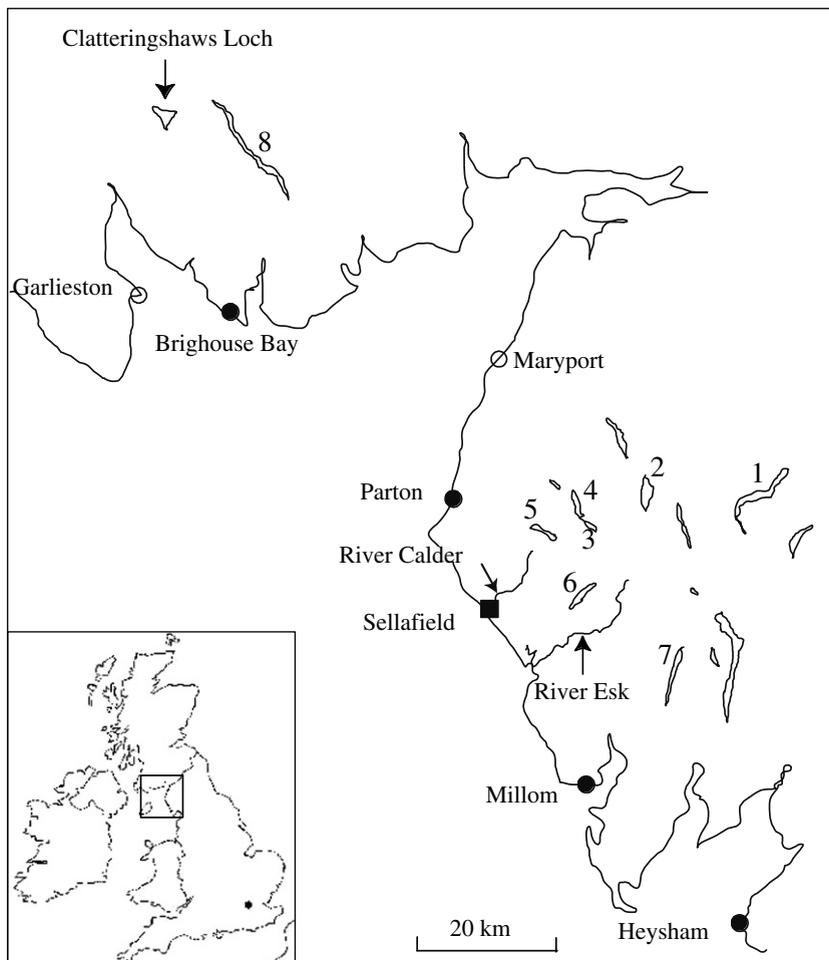


Fig. 2. Sampling points. (1) Ullswater, (2) Derwent Water, (3) Buttermere, (4) Crummock Water, (5) Ennerdale Water, (6) West Water, (7) Coniston Water, and (8) Loch Ken Closed circles show sampling point of seawater. River water was collected at 3.75 km from seashore of River Calder and at 8.75 km from seashore of River Esk.

Table 2
Distance from Sellafield and the sea to the middle point of the lakes

Sampling point	Distance from Sellafield (km)	Distance from the sea (km)
Ullswater	42.5	42
Derwent Water	28	27.5
Buttermere	19	20.5
Crummock Water	19.5	18
Ennerdale Water	13	12.5
Wast Water	13	13
Coniston Water	29	12.5
Loch Ken	25	74

organic I compounds into I^- , samples were oxidized with 5 mL 2.5% $NaClO_3$ solution for more than 15 min and reduced with $Na_2S_2O_5$ and $NH_2OH \cdot HCl$ at pH 5–6. The samples were acidified with HNO_3 and $NaNO_2$ was added to oxidize the I^- to I_2 in a separating funnel. Iodine was extracted into chloroform and back-extracted into water after reduction with $Na_2S_2O_5$. Then, the aqueous fraction was reduced in volume to about 30 mL by evaporating at 85–95 °C, followed by precipitation of $BaSO_4$ by adding saturated $Ba(NO_3)_2$ solution in order to minimize concentrations of S and associated Te. Iodide in the supernatant solution was precipitated as AgI by adding $AgNO_3$ solution. The AgI was dried at 70 °C and mixed with Ag . The $^{129}I/^{127}I$ ratio in this material was measured using AMS at SUERC (Maden et al., 2007) and ETH Zurich (López-Gutiérrez et al., 2000). Blanks were made by using the same stable I carrier, reagents and methods for water samples. The typical $^{129}I/^{127}I$ ratio of the blanks was 10^{-12} and the detection limit was 10^{-13} . The samples produced $^{129}I/^{127}I$ ratios of 10^{-10} – 10^{-9} . The Z94-0594 material provided by PRIME Laboratory at Purdue University was used as a standard for AMS measurements at SUERC, and this standard material was found to agree within 5% with a dilution of the NIST 4949B standard reference material (D2New), which was used as primary standard at ETH Zurich. The $^{129}I/^{127}I$ ratios of the original samples were derived from the AMS results for ^{129}I and measurement of ^{127}I concentrations by ICP-MS.

2.3. Analysis of ^{127}I

Filtered, alkaline sub-samples for ^{127}I analysis were diluted to an approximate I concentration of

1–2 $\mu g/kg$ using 18 M Ω water and a matrix solution that contained TMAH (tetramethyl-ammoniumhydroxide), $NaHSO_3$ and $NH_2OH \cdot HCl$ with concentrations in the sample of 0.08%, 0.01 M and 0.02 M, respectively. Then, Cs was added as the internal standard. Measurement of stable I was carried out using ICP-MS (PlasmaQuad II STE, VG Elemental). A standard solution was measured before and after every sample injection for calibration. Variation of counts by signal drift was corrected using count rates of Cs. Each sample solution was injected at least 3 times to get the average concentration and the relative uncertainty. The relative uncertainties of the concentrations were 4–16%.

3. Results and discussion

3.1. ^{127}I concentration

Table 3 shows the concentrations of ^{127}I and ^{129}I , and the $^{129}I/^{127}I$ ratio in water samples including the data for seawater samples in 1992 measured by Raisbeck et al. (1995). The major source of ^{127}I in the terrestrial environment is onshore transport from the sea. This is illustrated by the work of Steinnes and Frontasyeva (2002) who measured the concentrations of halogens (Cl, Br and I) in soil and showed that concentrations of all 3 elements decreased exponentially as a function of the distance from the ocean. This indicates that atmospheric supply from the marine environment is the predominant source of these elements in soil. However, in the present work, ^{127}I concentrations in lakes in the Lake District showed no correlation with the distance from the seashore (Fig. 3). Both Wast Water and Ennerdale Water, among the nearest lakes to the sea, showed lower ^{127}I concentrations, probably because of the geological characteristics of the catchment areas, such as soil organic matter content, pH and redox conditions. Peat soils generally have high I concentrations in zones of high organic matter content because of the ability of organic matter to retain I (Whitehead, 1984; Fuge and Johnson, 1986). The peat soil within the catchment areas of Wast Water and Ennerdale Water may prevent the flow of I into the lakes, and probably causes the low I concentration in the lake water. Given the presence of acid streams in the catchment areas of Wast and Ennerdale Waters (British Geological Survey, 1992), it is also possible that iodine is lost to the atmosphere due to volatilization following the oxidation of I^- to I_2 .

Table 3
 ^{127}I and ^{129}I concentrations, and $^{129}\text{I}/^{127}\text{I}$ ratios in water samples

Location	Sampling date	^{127}I ($\mu\text{g}/\text{kg}$)	^{129}I (at/kg)	$^{129}\text{I}/^{127}\text{I}$
<i>Lake</i>				
Ullswater	Mar 04	1.62 ± 0.21	$(9.05 \pm 1.21) \times 10^9$	$(1.18 \pm 0.04) \times 10^{-6}$
	Nov 04	3.07 ± 0.05	$(1.01 \pm 0.03) \times 10^{10}$	$(6.91 \pm 0.19) \times 10^{-7}$
Derwent Water	Mar 04	1.82 ± 0.11	$(1.19 \pm 0.10) \times 10^{10}$	$(1.37 \pm 0.07) \times 10^{-6}$
	Nov 04	3.17 ± 0.24	$(1.46 \pm 0.12) \times 10^{10}$	$(9.68 \pm 0.21) \times 10^{-7}$
Buttermere	Nov 04	1.59 ± 0.05	$(1.60 \pm 0.06) \times 10^{10}$	$(2.12 \pm 0.04) \times 10^{-6}$
	May 05	1.15 ± 0.05	$(2.37 \pm 0.05) \times 10^{10}$	$(4.34 \pm 0.19) \times 10^{-6}$
Crummock Water	Mar 04	2.30 ± 0.09	$(2.86 \pm 0.25) \times 10^{10}$	$(2.63 \pm 0.21) \times 10^{-6}$
	Nov 04	2.11 ± 0.05	$(3.38 \pm 0.10) \times 10^{10}$	$(3.38 \pm 0.06) \times 10^{-6}$
	May 05	1.54 ± 0.06	$(2.87 \pm 0.06) \times 10^{10}$	$(3.93 \pm 0.18) \times 10^{-6}$
Ennerdale Water	Nov 04	1.63 ± 0.03	$(4.38 \pm 0.13) \times 10^{10}$	$(5.67 \pm 0.13) \times 10^{-6}$
	May 05	1.23 ± 0.05	$(3.50 \pm 0.07) \times 10^{10}$	$(6.00 \pm 0.27) \times 10^{-6}$
Wast Water	Mar 04	0.91 ± 0.17	$(2.11 \pm 0.54) \times 10^{10}$	$(4.88 \pm 0.86) \times 10^{-6}$
	Nov 04	1.60 ± 0.06	$(2.99 \pm 0.15) \times 10^{10}$	$(3.94 \pm 0.14) \times 10^{-6}$
	May 05	1.24 ± 0.05	$(3.75 \pm 0.08) \times 10^{10}$	$(6.38 \pm 0.04) \times 10^{-6}$
	Jun 05	1.23 ± 0.05	$(3.37 \pm 0.07) \times 10^{10}$	$(5.77 \pm 0.04) \times 10^{-6}$
Coniston Water	Nov 04	3.13 ± 0.16	$(2.82 \pm 0.18) \times 10^{10}$	$(1.90 \pm 0.07) \times 10^{-6}$
Loch Ken	Nov 04	4.95 ± 0.19	$(5.82 \pm 0.28) \times 10^9$	$(2.48 \pm 0.07) \times 10^{-7}$
<i>River</i>				
River Calder	Nov 04	5.40 ± 0.20	$(2.11 \pm 0.09) \times 10^{11}$	$(8.25 \pm 0.19) \times 10^{-6}$
	May 05	3.20 ± 0.13	$(2.39 \pm 0.05) \times 10^{10}$	$(1.58 \pm 0.07) \times 10^{-6}$
River Esk	Nov 04	2.23 ± 0.13	$(4.55 \pm 0.28) \times 10^{10}$	$(4.30 \pm 0.09) \times 10^{-6}$
	May 05	1.46 ± 0.06	$(1.66 \pm 0.03) \times 10^{10}$	$(2.40 \pm 0.11) \times 10^{-6}$
Brook into Wast Water	Jun 05	2.83 ± 0.11	$(5.91 \pm 0.24) \times 10^{10}$	$(4.40 \pm 0.25) \times 10^{-6}$
River out of Wast Water	Jun 05	1.25 ± 0.05	$(3.17 \pm 0.06) \times 10^{10}$	$(5.35 \pm 0.24) \times 10^{-6}$
<i>Sea</i>				
Brighthouse Bay	Dec 04	25.1 ± 1.7	$(3.31 \pm 0.24) \times 10^{11}$	$(2.78 \pm 0.07) \times 10^{-6}$
Parton	Mar 04	33.5 ± 1.9	$(1.28 \pm 0.26) \times 10^{12}$	$(8.08 \pm 1.56) \times 10^{-6}$
Millom	Jun 05	32.7 ± 1.3	$(8.75 \pm 0.18) \times 10^{11}$	$(5.64 \pm 0.25) \times 10^{-6}$
Heysham	Dec 04	26.5 ± 2.0	$(1.03 \pm 0.08) \times 10^{12}$	$(8.20 \pm 0.19) \times 10^{-6}$
	Apr 92 ^a	36.5 ± 1.8	$(1.54 \pm 0.22) \times 10^{11}$	$(8.90 \pm 1.20) \times 10^{-7}$
Maryport ^a	Apr 92	36.2 ± 1.8	$(8.75 \pm 1.12) \times 10^{10}$	$(5.10 \pm 0.60) \times 10^{-7}$

^a From Raisbeck et al., 1995.

The ^{127}I concentration in the Irish Sea was reported by Truesdale (1994). They determined an I concentration of about $50 \mu\text{g}/\text{kg}$ offshore, decreasing to $42\text{--}43 \mu\text{g}/\text{kg}$ with decreasing distance from the seashore. In the present research, the ^{127}I concentration in seawater ($25\text{--}34 \mu\text{g}/\text{kg}$) was lower than observed by Truesdale (1994). The seawater was affected by the river flow, since the sampling points were close to inlets. The results were similar to the ^{127}I concentrations measured by Raisbeck et al. (1995) for seawater from Heysham and Maryport (16 km NW of Parton), with concentrations of

36.6 and $36.2 \mu\text{g}/\text{kg}$, respectively. Both freshwater and seawater ^{127}I concentrations are influenced by several factors, and the $^{129}\text{I}/^{127}\text{I}$ ratio is therefore probably a better tracer than the ^{129}I concentration to trace the transfer of ^{129}I released from nuclear facilities.

3.2. $^{129}\text{I}/^{127}\text{I}$ ratio in seawater

The $^{129}\text{I}/^{127}\text{I}$ ratio in seawater at Brighthouse Bay is lower than that at Parton, Millom and Heysham. Schnabel et al. (2007) measured the $^{129}\text{I}/^{127}\text{I}$ ratio

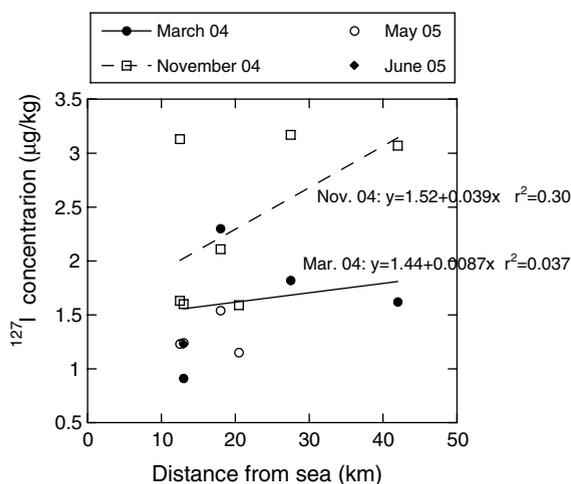


Fig. 3. ^{127}I concentration against distance from seashore.

in coastal water close to Brighthouse Bay (Garlieston; 15 km west of Brighthouse Bay) in 2005, and obtained a similar value (3.4×10^{-6}). This is consistent with the pattern of dilution and dispersion observed for soluble radionuclides discharged from Sellafield. Raisbeck et al. (1995) reported the $^{129}\text{I}/^{127}\text{I}$ ratios at Heysham and Maryport and these values were lower by one order of magnitude than those for Parton and Heysham in the present research, consistent with the increase in the ^{129}I content of the liquid effluent discharge from Sellafield over the last decade.

3.3. $^{129}\text{I}/^{127}\text{I}$ ratio in lake and river water

Loch Ken is 74 km distant from Sellafield and 25 km from the nearest seashore, so it is expected to be only slightly affected by the gaseous release from Sellafield. Thus, the $^{129}\text{I}/^{127}\text{I}$ ratio was expected to be similar to that of the sea around it. However, the observed $^{129}\text{I}/^{127}\text{I}$ ratio for Loch Ken water of 2.48×10^{-7} was one order of magnitude lower than that for the nearest seawater sample (Brighthouse Bay). Schnabel et al. (2007) also obtained the $^{129}\text{I}/^{127}\text{I}$ ratio (4.68×10^{-7}) at Clatteringshaws Loch, which is a smaller lake 20 km distant from a small bay fronting onto the Irish Sea and farther than Loch Ken from Sellafield (86 km). Iodine is released from the sea surface to the atmosphere as sea spray or as volatile I. Average atmospheric residence times are estimated to be 10 days for gaseous inorganic I, 18 days for gaseous organic I and 14 days for particulate I (Rahn et al., 1976). These atmospheric residence times

are sufficiently long for I to travel far away from the point of emission. Thus, long range transport of marine derived I with low $^{129}\text{I}/^{127}\text{I}$ ratio, from another part of the marine system (for example, outside the Irish Sea), could possibly cause the $^{129}\text{I}/^{127}\text{I}$ ratio to be lower in Loch Ken than in the nearest seawater sample. Another possible reason is the time delay in the terrestrial transfer of I from deposition on the soil or plant surface in the catchment area to inflow to the lake. The $^{129}\text{I}/^{127}\text{I}$ ratio in Irish Sea water has increased over the last decade in response to increasing ^{129}I content of the Sellafield liquid effluent discharge. Thus the I observed in Loch Ken may have been deposited before the increase in the liquid effluent discharge. Assuming that seawater is the dominant source of ^{129}I , the $^{129}\text{I}/^{127}\text{I}$ ratio in the Lake District would be lower than that in seawater in the Irish Sea for the same reason.

Fig. 4 shows the $^{129}\text{I}/^{127}\text{I}$ ratio in lake and river water against the distances from seashore and from Sellafield. The centres of Ullswater, Derwent Water, Buttermere, Crummock Water, Ennerdale Water and West Water, and the sampling points of the River Calder and River Esk are almost equidistant from both the seashore and the Sellafield nuclear fuel reprocessing plant. The centre of Coniston Water is 12.5 km from seashore and 29 km from

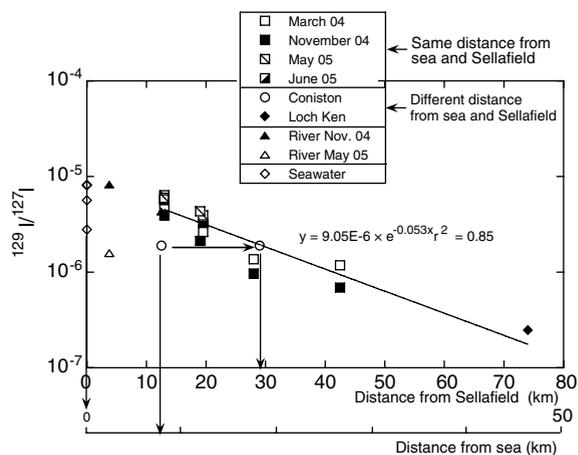


Fig. 4. $^{129}\text{I}/^{127}\text{I}$ ratio in lake and river water and in seawater against distance from seashore or the Sellafield nuclear reprocessing plant. Ullswater, Derwent Water, Buttermere, Crummock Water, Ennerdale Water, West Water, and the sampling point of River Calder and River Esk were almost equidistant from both the seashore and the Sellafield nuclear fuel reprocessing plant. The line is a regression line for the $^{129}\text{I}/^{127}\text{I}$ in lakes against distance from Sellafield.

Sellafield. The $^{129}\text{I}/^{127}\text{I}$ ratios in surface water collected from the Lake District varied from 6.91×10^{-7} to 8.25×10^{-6} . The $^{129}\text{I}/^{127}\text{I}$ ratios in the surface water were the same as or below the level of seawater at Parton, Millom and Heysham, and the $^{129}\text{I}/^{127}\text{I}$ ratios in lakes including Loch Ken decreased exponentially as a function of the distance from Sellafield ($r^2 = 0.85$). The $^{129}\text{I}/^{127}\text{I}$ ratio in the River Calder water in May 2005 was lower by a factor of 5 relative to that observed in November 2004 and was also significantly lower than the range of values observed for seawater. The $^{129}\text{I}/^{127}\text{I}$ ratio in the River Esk also decreased to almost half from November 2004 to May 2005. This is caused by decrease of ^{129}I concentration in these samples, probably correlating with decreased gaseous release from Sellafield, because river water is expected to respond rapidly to the change of ^{129}I deposition in the basin.

3.4. Relative contributions of liquid and gaseous ^{129}I emissions

The Sellafield gaseous discharges and onshore transport from the sea are both possible mechanisms of supply of ^{129}I to the terrestrial surface water in the Lake District. The $^{129}\text{I}/^{127}\text{I}$ ratio in Coniston Water, located close to two very broad intertidal areas, which could play an important role in the formation of methyl iodide and I_2 for the sea to air transfer of I, was 1.90×10^{-6} , twice that of Derwent Water, which is almost the same distance from Sellafield as Coniston Water but further from the seashore. The effect of ^{129}I from the Irish Sea probably caused the difference in the $^{129}\text{I}/^{127}\text{I}$ ratio between Coniston and Derwent Waters. However, consideration of the following relative variation of stable I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios suggests that Sellafield gaseous discharges also strongly affect the ^{129}I concentration in the lakes. The distance from seashore to West Water, Ennerdale Water and Coniston Water were almost the same, but the levels of $^{129}\text{I}/^{127}\text{I}$ in West Water and Ennerdale Water were, respectively, 2 and 3 times higher than that in Coniston Water, because West Water and Ennerdale Water were closer to Sellafield and hence receive more ^{129}I originating from Sellafield gaseous discharges. For Ullswater, Derwent Water and West Water, the $^{129}\text{I}/^{127}\text{I}$ ratio in November was lower than that in March, and conversely, for Crummock Water, the $^{129}\text{I}/^{127}\text{I}$ ratio in March was lower than that in November. This is because the

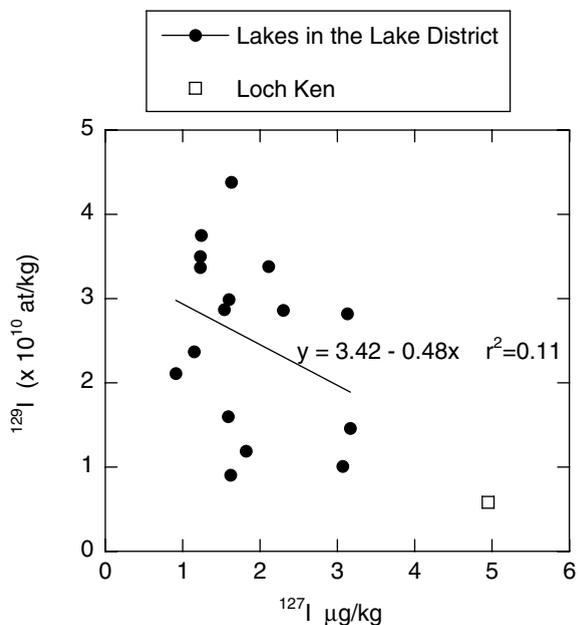


Fig. 5. ^{129}I concentration against ^{127}I concentration in lakes.

^{129}I concentration in the former 3 lakes increased in November but slightly decreased in Crummock Water, and the ^{129}I concentration increased (1.1–1.4 times) in all lakes. This different trend between the concentrations of ^{127}I and ^{129}I indicates that they have different sources. Fig. 5 shows that there is no correlation between ^{129}I and ^{127}I concentrations in lakes and the correlation r^2 value is 0.11.

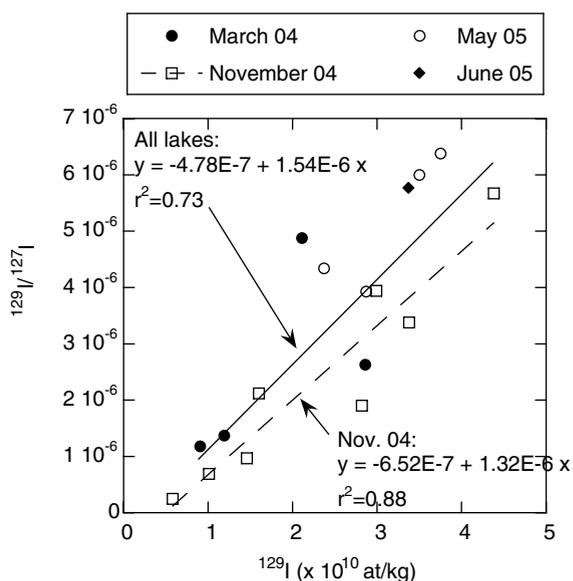


Fig. 6. $^{129}\text{I}/^{127}\text{I}$ ratio against ^{129}I concentration in lakes.

Fig. 6 shows the $^{129}\text{I}/^{127}\text{I}$ ratio against ^{129}I concentrations. The $^{129}\text{I}/^{127}\text{I}$ ratio increased linearly as ^{129}I concentration increased and the correlation r^2 value is 0.73 for all data of lakes and 0.88 for the lake samples collected in November 2004, the time of the most comprehensive sampling. These results indicate that the most likely dominant source of ^{129}I is Sellafield gaseous discharges, assuming that the steep increase of ^{129}I concentration in seawater has not affected the ratios. Furthermore, Rucklidge et al. (1994) found that the $^{129}\text{I}/^{127}\text{I}$ ratio in moss in the Lake District, at a time when the $^{129}\text{I}/^{127}\text{I}$ ratio in seawater was almost 10 times lower than now, ranged from 1.5×10^{-7} to 6.7×10^{-5} . These ratios are similar to the present results except for the moss collected from very close to the nuclear reprocessing plant, although the ratio in moss was not dependent on the distance from Sellafield.

4. Conclusions

$^{129}\text{I}/^{127}\text{I}$ ratios in the Irish Sea have increased by one order of magnitude from 1992 to 2004, corresponding to the increase in the ^{129}I content of liquid effluent discharges from Sellafield over the last decade. The $^{129}\text{I}/^{127}\text{I}$ ratios in lake water in the Lake District decreased exponentially as a function of distance from Sellafield. The Sellafield gaseous discharges and onshore transport from the sea are both possible mechanisms of supply of ^{129}I to the lakes. However, consideration of the relative variation of stable I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios suggests that Sellafield gaseous discharges may be the dominant source of ^{129}I to the lakes.

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