Supporting Information:

Estimate of European ¹²⁹I releases supported by ¹²⁹I analysis in an Alpine ice core

Herbert Reithmeier^{*a*,*}, Vitali Lazarev^{*a*}, Werner Rühm^{*b*}, Margit Schwikowski^{*c*,*d*}, Heinz W. Gäggeler^{*c*,*d*}, Eckehart Nolte^{*a*}

^aPhysics Department E15, TU München, D-85748 Garching, Germany

^bInstitute of Radiobiology, LMU München, D-80336 Munich, Germany

 $^{c}\mathrm{Laboratory}$ for Radiochemistry and Environmental Chemistry, PSI, CH-5232 Villigen, Switzerland

^dDepartment for Chemistry and Biochemistry, University of Bern, CH-3012 Bern, Switzerland

reactor	power $[MW_{th}]$	1950	1951	1952	1953	1954	1955	1956	1957
Windscale Pile 1	180	0.1	0.5	0.8	1.6	1.6	1.6	1.6	1.2
Windscale Pile 2	180		0.2	0.8	1.6	1.6	1.6	1.6	1.2
Calder Hall A	268							0.1	1.2
Calder Hall B	268								0.6
¹²⁹ I arise in repr.			0.3	1.3	2.4	3.2	3.2	3.2	2.8
reactor	power $[MW_{th}]$	1958	1959	1960	1961	1962	1963	1964	1965
Calder Hall A	268	2.4	2.4	2.4	2.4	2.4	2.4	2.4	2.4
Calder Hall B	268	1.2	2.4	2.4	2.4	2.4	2.4	2.4	2.4
Calder Hall C	268		0.6	1.2	2.4	2.4	2.4	2.4	2.4
Calder Hall D	268		0.6	1.2	2.4	2.4	2.4	2.4	2.4
Chapelcross A-D	4 × 948		17	39	88	88	8.8	8.8	8.8
-	4 × 240		1.1	0.0	0.0	0.0	0.0	0.0	
Berkeley A+B	4×248 2×556		1.1	0.0	0.0	2.5	5.0	9.9	9.9
Berkeley A+B Bradwell A+B	2×556 2×531		1.1	0.0	0.0	2.5 2.5	$5.0 \\ 5.0$	9.9 9.9	9.9 9.9

Estimation of ¹²⁹I emissions from fuel reprocessing at Windscale/Sellafield

TABLE 1: Estimated annual production of ¹²⁹I [GBq] in the British Magnox reactors. The arisen ¹²⁹I [GBq], i.e. the ¹²⁹I present in spent fuel at the time of its reprocessing, at Windscale/Sellafield is estimated from the ¹²⁹I produced in the Magnox reactors. The thermal powers and start of operation of the considered Magnox reactors are taken from [1, 2].

^{*}Corresponding author e-mail: reithmei@ph.tum.de

reactor	power $[MW_{th}]$	periode of operation	mean production of 129 I [GBq/y]	total ¹²⁹ I produced [GBq]
Marcoule G1	40	1956 - 68	0.4	4.7
Marcoule G2	200-250	1958 - 80	2.0	45.8
Marcoule G3	200-250	1959 - 84	2.1	53.9
Chinon-1	300	1963 - 73	2.5	28.0
Chinon-2	848	1965 - 85	7.2	152
Chinon-3	1560	1966 - 90	11.5	287
Célestin $1+2$	2×200	1967 - 91	2.2	53.9
St.Lorant-A1	1652	1969 - 90	12.7	279
St.Lorant-A2	1700	1971 - 92	12.7	279
Bugey-1	1950	1972 - 94	14.6	336
Phénix	563	1973 - 89	4.4	74.9
Vandellos-1	1600	1972 - 90	2.4	47.0
sum				1654

Estimation of ¹²⁹I emissions from fuel reprocessing at Marcoule

TABLE 2: Estimated mean and total ¹²⁹I production at reactors, from which spent fuel was reprocessed at Marcoule [3]. The estimation is based on their thermal powers and the periods of operation, taken from [1, 3, 4]. The thermal power of the Vandellos-1 reactor (Spain) is estimated from its electrical power of 480 MW_e [4].

year								1958	1959	1960
129 I [GBq]								0.2	0.2	1.3
year	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970
¹²⁹ I [GBq]	3.6	4.0	4.0	4.0	3.7	4.2	7.6	8.5	8.5	11.5
year	1971	1972	1973	1974	1975	1976	1977	1978	1979	1980
¹²⁹ I [GBq]	8.9	9.5	9.5	10.7	26.8	52.7	75.8	75.7	75.7	75.7
year	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990
129 I [GBq]	75.7	74.6	73.5	84.4	79.9	76.7	73.4	65.7	65.7	65.7
year	1991	1992	1993	1994	1995	1996	1997			
$^{129}I [GBq]$	65.7	61.1	47.9	31.3	31.3	17.8	16.5			

TABLE 3: Estimated ¹²⁹I arisen in reprocessing at the Marcoule plants UP-1 and APM.

Estimation of ¹²⁹I re-emission from the ocean

The tropospheric stable iodine concentration can be explained by a continual source from the oceans. To maintain the constant stable iodine concentration in air of 3.5 ppt by weight [5], the deposition rate \dot{n}_{\downarrow} to and the emission rate \dot{n}_{\uparrow} from the oceans must be equal.

Supposing that the emission rate of stable iodine, \dot{n}_{\uparrow} , scales with the concentration in the ocean and with the area of the ocean surface, a rate constant v_V for ¹²⁹I can be deduced (equation 1)

$$\dot{n}_{\uparrow} = c_o^I \times A_o \times v_V \approx \dot{n}_{\downarrow},\tag{1}$$

where \dot{n}_{\uparrow} is the emission rate of stable iodine from the ocean, \dot{n}_{\downarrow} is the deposition rate of stable iodine to the ocean, $c_o^I = 47 \,\mu \text{g/l}$ is the concentration of stable iodine in the ocean [5], and A_o is the ocean surface. The rate constant v_V can now be interpreted as the depth h_{\uparrow} , of which the complete iodine inventory has to be emitted from the ocean's surface to maintain a constant concentration of stable iodine in air.

The re-emission rate of ¹²⁹I from the ocean, \dot{N}_{\uparrow} , was calculated the same way, assuming wellmixed ¹²⁹I and ¹²⁷I as well as evaporation of all the ¹²⁹I contained in the water layer down to the depth h_{\uparrow} (equation 2):

$$\dot{N}_{\uparrow} \approx N_M \times \frac{v_V}{\bar{h}}.$$
 (2)

Here, N_M is the ¹²⁹I inventory in the ocean water with the corresponding mean depth \bar{h} . We assumed a mean depth of 50 m for the North Sea, and of 150 m for the Norwegian Sea and the Barents Sea, respectively.

The transport time of 129 I from the site of liquid release to the region of deep water formation close to Greenland, i.e. the residence time in the Atlantic, was estimated as follows [6, 7]:

- The transport time from La Hague to West Spitsbergen through the North Sea and along Norway is 4 y. 61 % of the Gulf Stream take this way.
- Transport from La Hague through the North Sea and the Barents Sea and back through the Fram Strait to the region of deep water formation close to Greenland takes 12 y. 39% of the Gulf Stream take this way.
- The transport time from Sellafield (Irish Sea) to the North Sea around Scotland is 2 a. From there on, the transport times are the same like for La Hague, so that the transport times from Sellafield to the region of deep water formation close to Greenland each are 2 years longer than the transport times from La Hague.

For the estimation presented here, a residence time for iodine in air of 15 d was used, based on the typical time for decomposition of methyl iodide of $t_d=3$ d [5], which is one of the major chemical forms of iodine in air, and on the tropospheric residence time t_r of the produced iodide, which we estimated to be $t_r=12$ d.

Year	sample	depth	129 I conc.	¹²⁹ I dep.
	[ml]	[m we]	$[10^{8} at/l]$	$[10^{12} at/m^2 y]$
1970/71	43	64.2	1.21 ± 0.19	0.19 ± 0.03
1972/73	69	61.0	0.58 ± 0.16	0.15 ± 0.04
1974	31	55.7	1.47 ± 0.10	0.24 ± 0.02
1975/76	35	54.1	0.61 ± 0.31	0.10 ± 0.05
1977/78	54	51.0	3.04 ± 0.50	0.70 ± 0.12
1979/80	42	46.3	2.12 ± 0.47	0.39 ± 0.09
1981	27	42.6	0.54 ± 0.20	0.08 ± 0.03
1982	21	41.1	1.44 ± 0.26	0.16 ± 0.03
1983	20	40.0	10.8 ± 0.44	1.85 ± 0.08
1984	26	38.3	6.11 ± 1.18	0.87 ± 0.17
1985	34	36.9	1.63 ± 0.76	0.25 ± 0.12
1986	19	35.3	6.34 ± 2.53	0.61 ± 0.24
1987	47	34.4	8.39 ± 1.25	1.82 ± 0.27
1988	41	32.2	4.58 ± 0.57	0.90 ± 0.11
1989/90	39	28.2	1.65 ± 0.24	0.33 ± 0.05
1991	43	26.3	1.27 ± 0.22	0.24 ± 0.04
1992/93	56	22.8	0.81 ± 0.27	0.15 ± 0.05
1994/95	49	18.9	2.79 ± 0.41	0.48 ± 0.07
1996	31	17.3	2.27 ± 0.57	0.45 ± 0.11
1997	31	15.3	1.23 ± 0.17	0.37 ± 0.05
1998	26	12.3	1.11 ± 0.29	0.27 ± 0.07
1999/00	52	7.4	0.77 ± 0.14	0.18 ± 0.03
2001/02	55	2.7	1.23 ± 0.21	0.32 ± 0.06

Results of ¹²⁹I analysis at Fiescherhorn glacier

TABLE 4: Sample volume, depth of the lower horizon of the considered ice layer in meter water equivalent (m we), measured ¹²⁹I concentrations, and resulting ¹²⁹I deposition fluxes.

Comparison with ¹²⁹I concentrations in rain water

Rain water, Dübendorf	129 I conc. [10 ⁸ at/l]	129 I dep. flux [10^{12} at/m ² y]	Fiescherhorn glacier	¹²⁹ I dep. flux [10 ¹² at/m ² y]
02/94-12/94	31.6	3.8	1994/95	0.48
01/95-09/95 08/96-06/97	$26.3 \\ 33.1$	$\frac{2.7}{1.7}$	1996 1997	$\begin{array}{c} 0.45\\ 0.37\end{array}$

TABLE 5: ¹²⁹I concentrations in rain water from Dübendorf and resulting ¹²⁹I deposition fluxes [8], compared with the corresponding data from the Fiescherhorn glacier.

References and Notes

- (1) Commission of the European Communities. Radioactive effluents from nuclear power stations and nuclear fuel reprocessing plants in the Community, discharge data 1969-1974. Commission of the European Communities Report. Luxembourg. 1975.
- (2) Bellona Foundation. Working paper 5: Sellafield. Bellona Foundation, http://www.bellona.no, Oslo. 2001.
- (3) Barrillot, B.; Davis, M. Les déchets nucléaires militaires français. Centre de Documentation et de Recherche sur la Paix et les Conflicts (CRDS). Lyon. 1994.
- (4) Stricht, S.V.; Janssens, A. Eds. Radiation Protection 127. Radioactive effluents from nuclear power stations and nuclear fuel reprocessing plants in the European Union, 1995-99. European Commission. Luxembourg. 1999.
- (5) Vogt, R. Iodine Compounds in the Atmosphere. In: *The Handbook of Environmental Chemistry Vol.4 Part E.* Springer-Verlag. Berlin-Heidelberg. 1999.
- (6) Nies, H.; Harms, I.H.; Karcher, M.J.; Dethleff, D.; Bahe, C.; Kuhlman, G.; Kleine, E.; Loewe, P.; Oberhuber, J.M.; Backhaus, J.O.; Matishov, D.; Stepanov, A.; Vasiliev, O.F. Anthropogenic radioactivity in the Arctic Ocean: Results from a joint project. *Germ. J. Hydrography* 1998, 50(4), 313-343.
- (7) Karcher, M.J.; Oberhuber, J.M. Pathways and modification of the upper and intermediate waters of the Arctic Ocean. J. Geophys. Res. 2002, 107(C6), 3049.
- (8) Schnabel, C.; Lopez-Gutierrez, J.M.; Szidat, S.; Sprenger, M.; Wernli, H.; Beer, J.; Synal, H.-A. On the origin of ¹²⁹I in rain water near Zürich. *Radiochimica Acta* 2001, 89, 815-822.