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The Potential Of I-129 as an Environmental Tracer

Andrej Osterc and Vekoslava Stibilj
*Institute Jožef Stefan,
Slovenia*

1. Introduction

Iodine has two natural isotopes – the only stable iodine isotope is ^{127}I , whilst ^{129}I is the only radioactive iodine isotope that is formed in nature ($T_{1/2} = 1.57 \cdot 10^7$ years). However, the main sources of ^{129}I in the environment are anthropogenic from nuclear fuel reprocessing plants (NFRP) and nuclear accidents. Current levels of ^{129}I do not represent any radiological hazard to humans, but the liquid discharges of ^{129}I from reprocessing plants into the ocean makes it a unique oceanographic tracer to study the movement of water masses, transfer of radionuclides and marine cycles of stable elements such as iodine. The gaseous releases of ^{129}I from reprocessing plants can be used as an atmospheric and geochemical tracer (Hou, 2004).

^{129}I and ^{127}I have the same chemical properties and therefore it is expected that they also behave similar in environment. Lack of ^{129}I and ^{127}I speciation data makes it difficult to confirm or disprove this assumption. The main problem is the mobility – species of newly introduced and old – natural ^{129}I . The old ^{129}I is in equilibrium with ^{127}I – natural $^{129}\text{I}/^{127}\text{I}$ ratio and this is disturbed with ^{129}I from NFRP which is released to the environment in volatile form. As such it is rapidly transferred among surface compartments. Liquid discharges to oceans influence areas in accordance with marine currents. Wet and, to a lesser extent, dry depositions of atmospheric ^{129}I are the main sources for ^{129}I in terrestrial environment, which is distant from ^{129}I sources such as NFRP.

The biggest reservoir of iodine is the ocean with an average concentration of approximately $50\text{-}60 \mu\text{g L}^{-1}$ seawater. From marine environment is iodine transferred to the atmosphere by volatilization mainly as iodomethane (CH_3I) and then washed out to terrestrial environment by wet and dry deposition. It is accumulated in soils where it is strongly bound-adsorb to organic matter, and iron and aluminium oxides in soil (Fuge, 2005). In the accumulation processes of iodine in soil besides various physico-chemical parameters including soil type, pH, Eh, salinity, and organic matter content, soil microorganism – especially bacteria were found to play an important role (Muramatsu & Yoshida, 1999, Amachi, 2008). In this way the biogeochemical cycling of ^{129}I is strongly connected to processes in ocean and soil systems – the atmosphere being the bridge between them.

2. Sources, inventory and levels of ^{129}I in marine and terrestrial environment

All ^{129}I formed in the primordial nucleosynthesis decayed to stable ^{129}Xe . Two natural processes responsible for natural background levels of ^{129}I are spallation of cosmic rays on

atmospheric Xe (cosmogenic) in the upper atmosphere and spontaneous fission of ^{238}U (fissiogenic).

Although ^{129}I is produced naturally the main part is a consequence of human nuclear activities (Table 1). In this way the sources can be divided in natural and man-made or in pre-nuclear and nuclear era. From 1945 anthropogenic sources of ^{129}I were nuclear weapons testing, nuclear accidents (Chernobyl) and at present marine and atmospheric discharges from NFRP. Operating plants in Europe are located in England (Sellafield), France (La Hague) and Russia (Mayak), and outside Europe in China, India, Pakistan and Japan (Tokaimura, Rakkasho). ^{129}I is produced during the operation of a nuclear power reactor by nuclear fission of $^{235}\text{U}(n, f)^{129}\text{I}$ and $^{239}\text{Pu}(n, f)^{129}\text{I}$. It was estimated that about 7.3 mg of ^{129}I is produced per megawatt day. ^{129}I is released during reprocessing of nuclear fuel – mainly by PUREX process. The fuel is first dissolved with nitric acid and at this step iodine is oxidized to volatile I_2 and despite all efforts to trap and collect released iodine some part may be discharged from the NFRP (Reithmeir et al., 2006).

Source	Inventory/release (kg)**	$^{129}\text{I}/^{127}\text{I}$ ratio in environment
Nature	250	$\sim 1 \cdot 10^{-12}$
Nuclear weapons testing	57	$1 \cdot 10^{-11} - 1 \cdot 10^{-9}$
Chernobyl accident	1.3–6	$10^{-8} - 10^{-6}$ (in contaminated area)
Marine discharges from European NFRP* by 2007	5200	$10^{-8} - 10^{-6}$ (North Sea and Nordic Sea water)
Atmospheric releases from European NFRP* by 2007	440	$10^{-8} - 10^{-6}$ (in rain, lake and river water in West Europe) $10^{-6} - 10^{-3}$ (in soil, grass near NFRP)
Atmospheric releases from Hanford NFRP*	275	$10^{-6} - 10^{-3}$ (in air near NFRP)

*NFRP...nuclear fuel reprocessing plant; **Marine discharges are sum discharges from La Hague and Sellafield NFRP, Atmospheric releases are sum releases from La Hague, Sellafield, Marcoul and Karlsruhe-WAK (after Hou et al., 2009)

Table 1. Sources and $^{129}\text{I}/^{127}\text{I}$ ratio in environment

Until the beginning of the 1990s the total annual discharges from two European NFRP, La Hague and Sellafield, remained below 20 kg year^{-1} . The discharges increased later considerable – up to 300 kg year^{-1} and accounted until 2000 for more than 95 % of the total inventory in the global ocean (Fig. 1) (Alfimov et al., 2004; Lopez-Gutierrez et al., 2004).

The natural, pre-nuclear $^{129}\text{I}/^{127}\text{I}$ isotopic ratio was significantly influenced by releases of anthropogenic ^{129}I to the environment. The estimated pre-nuclear $^{129}\text{I}/^{127}\text{I}$ isotopic ratio in marine environment was assessed with analysis of marine sediments and agreed to be $1.5 \cdot 10^{-12}$ (Table 2) (Moran et al., 1998; Fehn et al., 2000a; Fehn et al., 2007). For the terrestrial environment – pedosphere and biosphere no agreed data on pre-nuclear ratio exist. Human nuclear activity increased the $^{129}\text{I}/^{127}\text{I}$ ratio in marine environment to $10^{-11} - 10^{-10}$ and to $10^{-8} - 10^{-5}$ (Table 3) in the Irish Sea, English Channel, North Sea and Nordic Seas which are influenced by liquid discharges from European NFRP (Frechou & Calmet, 2003; Alfimov et al., 2004; Hou et al., 2007). In the terrestrial environment the $^{129}\text{I}/^{127}\text{I}$ ratio increased to $10^{-9} -$

10^{-7} , even 10^{-6} – 10^{-4} in the vicinity of nuclear fuel reprocessing plants (Table 4) (Duffa & Frechou, 2003; Frechou & Calmet, 2003).

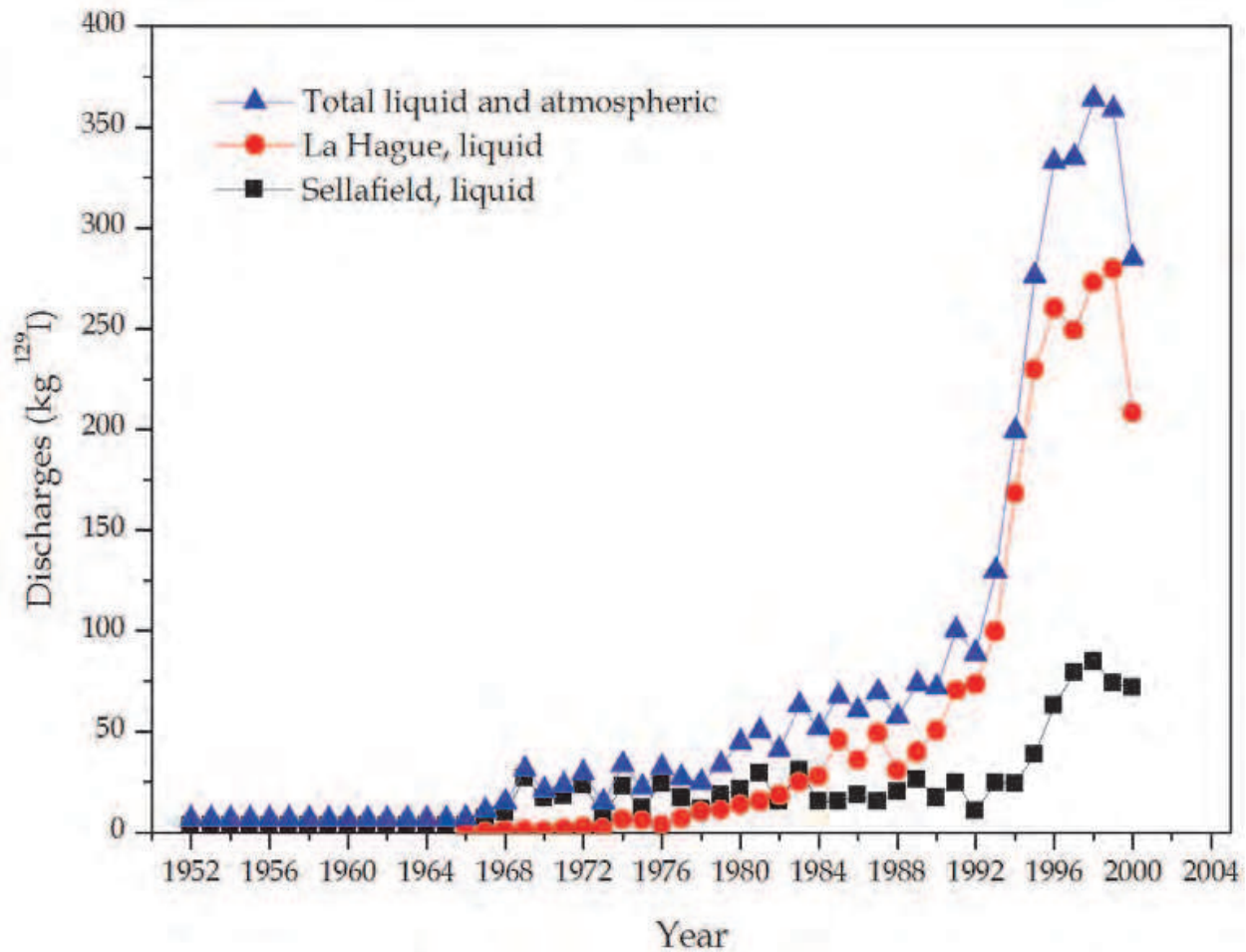


Fig. 1. Liquid and atmospheric releases of ^{129}I from NFRP in La Hague and Sellafield for period from 1952 to 2000 (compiled by Lopez-Gutierrez et al., 2004).

Atmospheric releases are not plotted, but they are considered in the total amount. Annual atmospheric releases ranged from 1.19 to 9.58 kg ^{129}I with a total amount of 235.5 kg in the period from 1952 to 2000.

Anthropogenic ^{129}I predominates in marine environment in biosphere and upper layers of the oceans and in terrestrial environment in soil, therefore it can be expected that the isotopic ratio $^{129}\text{I}/^{127}\text{I}$ is increasing in these compartments of the ecosystem. Precipitation and seawater are probably the main carriers for ^{129}I exchange among different compartments in marine and terrestrial environment. Data from literature clearly show that ^{129}I levels in marine sediment, marine algae and soil are several times higher than in seawater or precipitation. Meaning that ^{129}I is most probably chemically or biologically transformed to species which accumulate in those compartments (Tables 3 and 4).

To summarize, different values of $^{129}\text{I}/^{127}\text{I}$ isotopic ratios in environment are today envisaged as 10^{-12} for pre-nuclear era, 10^{-9} in slightly contaminated regions and 10^{-9} – 10^{-6} in regions affected by the releases from NFRP. The highest ratios were found in the close vicinity of NFRP with values from 10^{-6} to 10^{-4} (Hou, 2009).

Sample	$^{129}\text{I}/^{127}\text{I}$ (10 ⁻¹²)	Reference
<i>TERRESTRIAL ENVIRONMENT</i>		
<i>Soil</i>		
Russia, Moscow, 1910	168	Szidat et al., 2000a
Russia, Bogoroditsk, 1909	25	
Russia, Lutovinovo, 1939	5.7	
<i>Thyroid powder</i>		
Species not given (USA), 1947	4.6	Szidat et al., 2000a
USA, Pig, 1947	58	
USA, Horse, 1947	1230	
<i>MARINE ENVIRONMENT</i>		
<i>Sediment</i>		
Peru, depth: 155-199 cm	1.50	Moran et al., 1998
Mexico, Baja peninsula, depth: 415-420 cm	1.48	
Ecuador, depth: 315-320 cm	1.05	
<i>Algae</i>		
Japan <i>Laminaria Japonica</i>		Fehn et al., 2007
Hokkaido, 1883	1.40	
Hokkaido, 1883	0.55	
Miyagi, 1883	0.52	
Miyagi, 1883	0.67	
<i>Pelvita</i>		
Miyagi, 1904	1.87	
Russia <i>Laminaria digitata</i>		Cooper et al., 1998
Novaya Zemlya, 1930	1.00	
Novaya Zemlya, 1931	3.69	
White Sea, 1938	1.35	
White Sea, 1930	1.37	
White Sea, 1938	1.92	

Table 2. $^{129}\text{I}/^{127}\text{I}$ isotopic ratios in pre-nuclear age environmental and biological samples

3. Factors affecting biogeochemical cycling of iodine

Iodine is a trace element present in the hydrosphere, lithosphere, atmosphere and biosphere at different concentrations and as different iodine species (Table 5). Speciation analysis of iodine was mainly done on stable ^{127}I (Hou et al., 1997; dela Veija et al., 1997; Sanchez & Szpunar, 1999; Hou et al., 2000c; Leiterer et al., 2001; Schwehr & Santschi, 2003; Shah et al., 2005; Gilfedder et al., 2008), with some studies on ^{129}I (Hou et al., 2001; Hou et al., 2003b; Schwehr et al., 2005; Englund et al., 2010b). Majority of researches performed on ^{127}I and ^{129}I are limited to fractionations of iodine – water soluble, exchangeable, bound to oxides, organic-inorganic fraction, etc. In general just the most abundant chemical forms of iodine – iodide (I^-) and iodate (IO_3^-) are determined and the rest of total iodine content is associated with organic iodine. It is well known that organic iodine fraction mainly consist of iodine

Sample	$^{129}\text{I}/^{127}\text{I}$ (10^{-8})	Reference
<i>Sea water</i>		
Germany, North Sea, 1999	153	Szidat et al., 2000a
Greenland, 1999 (n = 5)	0.07–0.24	Hou, 2004
England, Irish Sea, near Sellafield 2004-05 (n = 4)	89–820	Atarashi-Andoh et al., 2007
Scotland, Scottish Sea, influence of Sellafield, 2003-2005 (n = 14)	7.2–336	Schnabel et al., 2007
Israel, Sea of Galilee, June 1998	0.31	Fehn & Snyder, 2000b
Israel, Engedi, Dead Sea, June 1998	0.003	
Japan Sea, Toyama Bay, October 2006	0.0086	Suzuki et al., 2008
Japan Sea, Off Sekine, 2006-2007 (n = 2)	0.0063–0.0068	
<i>Sediment</i>		
Sweden, Baltic Sea, influence of La Hague and Sellafield, core sample –from 0 to 21 cm, 1997	0.34–1.06	Aldahan et al. 2007
<i>Seaweed</i>		
Greenland, 1997 (<i>Fucus distichus</i> , n = 7)	0.07–0.15	Hou et al., 2000a
Norway (Utsira), 1980-1995 (<i>Fucus vesiculosus</i> , n = 16)	1.88–18.5	
Denmark (influence of liquid discharges from NFRP) Roskilde Fjord and Bornholm, 1995-1998 (<i>Fucus vesiculosus</i> , n = 8)	2.50–9.12	
Klint, 1986-1999 (<i>Fucus vesiculosus</i> , n = 39)	3.54–37.5	
France (vicinity of La Hague)		Frechou et al., 2003
Goury, 1998-1999 (<i>Fucus vesiculosus</i> , n = 3)	1010–1940	
Goury, 1998-1999 (<i>Fucus serratus</i> , n = 3)	930–1210	
Goury, 1998-1999 (<i>Laminaria digita</i> , n = 2)	540–1270	Barker et al., 2005
Goury and Dielette, 2003 (<i>Fucus serratus</i> , n = 12)	496–1960	
Goury and Dielette, 2003 (<i>Laminaria digita</i> , n = 8)	349–960	
Ireland		Keogh et al., 2007
West and South coastline, <i>Fucus vesiculosus</i>		
1985, n = 7	0.08–0.73	
1994, n = 7	0.47–6.5	
2003, n = 9	0.21–5.0	
East coastline (influence of liquid discharges from NFRP), <i>Fucus vesiculosus</i>		
1985, n = 8	4.8–85	
1994, n = 7	0.83–30	
2003, n = 8	24–85	
Russia, <i>Laminaria digitata</i>		Cooper et al., 1998
Murmansk region, 1966	0.016	
Murmansk region, 1967	0.034	
White Sea, 1971	0.027	
Novaya Zemlya, 1989	0.48	
Novaya Zemlya, 1993	0.72	
Slovenia, Adriatic Sea, <i>Fucus virsoides</i> , September 2005, five locations	0.086–0.11	Osterc & Stbilj, 2008
Italy, Adriatic Sea, <i>Fucus virsoides</i> , June 2006, five locations	0.068–0.15	
Croatia, Adriatic Sea, <i>Fucus virsoides</i> , October 2006, three locations	0.15–0.31	

Table 3. $^{129}\text{I}/^{127}\text{I}$ isotopic ratios in nuclear age environmental and biological samples from marine compartments

Sample	$^{129}\text{I}/^{127}\text{I}$ (10^{-8})	Reference
<i>Aerosol</i>		
Spain, Seville, 2001 (n = 12)	0.29–2.72	Santos et al., 2005
Spain, Seville, 2001-2002	0.18–5.35	Santos et al., 2006
Sweden, Kiruna and Ljungbyhed, 1983-2008	0.5–147	Englund et al., 2010a
<i>Gas</i>		
Spain, Seville, 1993-1994 and 1998	0.01–0.80	Santos et al., 2006
<i>Precipitation</i>		
Germany, Hanover, 1986	16.6	Szidat et al., 2000a
Germany, Lower Saxony, 1997	83.4	
Germany, Upper Bavaria, 2003	14.6–38.6	Reithmeier et al., 2005
Spain, Seville, 1996-1997	0.23–52	Santos et al., 2006
Antartica, McMurdo Station, snowmelt 1999	0.004	Snyder et al., 2004
Antartica, Mt Erebus, snow, 2000	0.009	
<i>Shallow ground water</i>		
Germany, Lower Saxony, 1997	0.8	Szidat et al., 2000a
<i>Lake water</i>		
Denmark, 2000 (n = 7)	2.5–27.3	Hou, 2004
Lithuania, 1999 (n = 2)	6.6–7.3	Hou et al., 2002
England, lakes near Sellafield, 2004-2005 (n = 7)	24.8–638	Atarashi-Andoh et al., 2007
Germany, Munich, Kleinhesselohrer See, July 1997	2.4	Fehn & Snyder, 2000b
Germany, Malchow, Malchow See, July 1997	8.6	
Germany, Harz, Okersee, June 1999	1.0	Snyder et al., 2004
USA, Oregon, Crater Lake, September 1996	0.9	
USA, Colorado, Navajo Lake, June 2000	0.25	Snyder et al., 2003a
Central America, Nicaragua, Lake Managua, 1998	0.029	Fehn & Snyder, 2000b
South America, Chile, Lago Verde, Februar 1999	0.24	Snyder et al., 2004
Australia, New South Wales, Lake George, 1997	0.53	Fehn & Snyder, 2000b
New Zealand, Lake Taupo, 1999	0.005	Snyder et al., 2004
Japan, Odanoike lake, May 2000	0.79	
Indonesia, Bali, Lake Beratan	0.032	
<i>River water</i>		
England, London, river Thames, March 1999	1.9	Snyder et al., 2004
England, Cambridge, river Granta, March 1999	1.0	
England, rivers near Sellafield 2004-2005 (n = 4)	158-825	Atarashi-Andoh et al., 2007
USA, Colorado, Pine River, June 2000	0.13	Snyder et al., 2003a
USA, Colorado, Animas River, June 2000	0.08	
India, Tista River, 1999	0.18	Snyder et al., 2004
India, Ganges River, 1999	0.03	
Central America, El Salvador, Rio Lempa, 1999	0.058	Snyder et al., 2003b
Africa, Botswana, Thamakkane river, May 2000	0.10	Snyder et al., 2004
Japan, Kugino river, May 2000	0.04	
Mongolia, Tuyu Gol River, January 2000	0.068	
<i>Thyroid</i>		
France, vicinity of La Hague (1–30 km), 1980-1999 (Bovine, n = 19)	100–25068*	Frechou et al., 2003
China (Tianjin), 1994-1995 (Human, male; n = 4)	0.04–0.09	Hou et al., 2000b
China (Tianjin), 1995 (Human, female; n = 2)	0.16–0.20	

*The highest isotopic ratio ($2.5 \cdot 10^4$) was obtained for an animal coming from Digulleville, a village 3 km to the north-east of the NFRP

Table 4. $^{129}\text{I}/^{127}\text{I}$ isotopic ratios in nuclear age environmental and biological samples from terrestrial compartments

Compartment	Main iodine species	Reference
	Concentration range	
Atmosphere	particle associated (aerosol); inorganic gaseous: I ₂ , HI, HIO; organic gaseous: CH ₃ I, CH ₂ I ₂ , CH ₃ CH ₂ CH ₂ I, etc.	Hou et al., 2009
	1-100 ng m ⁻³	Wershofen & Aumann, 1989; Yoshida & Muramatsu, 1995
<i>Hydrosphere</i>		
oceans	inorganic: I ⁻ , IO ₃ ⁻ ; organic: CH ₃ I	Hou et al., 2001
	45-60 ng mL ⁻¹	Hou et al., 2009
fresh water	1-3 ng mL ⁻¹	Hou et al., 2009
precipitation	1-6 ng mL ⁻¹	Yoshida & Muramatsu, 1995; Hou, 2004
<i>Lithosphere</i>		
soil	inorganic: I ⁻ , IO ₃ ⁻ , bound to metal oxides, carbonates and minerals; organic: bound to humic and fulvic acids	Schmitz & Aumann, 1995
	0.5-40 µg g ⁻¹	Muramatsu & Yoshida, 1999
surface sea sediment	1-2000 µg g ⁻¹	Muramatsu & Wedepohl, 1998
metamorphic and magmatic rocks	<0.1 µg g ⁻¹	
<i>Biosphere</i>		
seaweed	inorganic: I ⁻ , IO ₃ ⁻ ; organic: iodo-amino acids (<i>Laminaria japonica</i>); bound to proteins, pigments, polyphenols*	Hou et al., 1997 Hou et al., 2000c Shah et al., 2005
	10-6000 µg g ⁻¹	Hou & Yan, 1998; Osterc & Stibilj, 2008
plants (terrestrial)	<1 µg g ⁻¹	Hou et al., 2009
thyroid gland	inorganic: I ⁻ organic: iodo-amino acids → iodo-thyronine and iodo- tyrosine	dela Vieja et al., 1997
	500-5000 µg g ⁻¹	Hou et al., 2003a
milk (bovine)	inorganic: I ⁻ organic: bound to proteins*	Leiterer et al., 2001
	0.017-0.49 µg mL ⁻¹	

*species not identified

Table 5. Concentrations of stable iodine in environmental compartments

bound to proteins – but these are still not identified for most environmental and biological samples, not for ^{127}I and certainly not for ^{129}I . The main problem is lack of appropriate standards for speciation analysis and very small amounts of ^{129}I in environmental and biological samples.

Iodine is released from marine environment to the atmosphere partly as aerosols formed from the sea spray – inorganic iodide and iodate – and mainly as volatile organic iodine compounds (VOIC) such as iodomethane (Baker et al., 2000; Leblanc et al., 2006, Chance et al., 2009). Bacteria, phytoplankton and brown algae present in marine environment are capable to reduce the most thermodynamically stable form of iodine, the iodate to iodide. On the other hand microalgae and macroalgae-seaweed accumulate iodide and transform it into VOIC – the most important are CH_3I , CH_2I_2 , CH_2BrI and CH_2ClI (Leblanc et al., 2006). The emitted organic iodine is decomposed by sunlight into inorganic iodine compounds. The photolytic lifetimes of VOIC differ; CH_2I_2 has a lifetime of 5 minutes, followed by CH_2BrI with a lifetime of 45 minutes and CH_2ClI with a lifetime of 10 h (Stutz, 2000). The longest photolytic lifetime of 14–18 days has CH_3I (Stutz, 2000). During this process of photolization reactive iodine oxides such as HOI , I_2O_2 and IO_2 form, which either form condensable vapours as nuclei for aerosols or react with ozone. From the atmosphere iodine enters the marine and terrestrial environment by processes of wet and dry deposition. In the iodine terrestrial cycle interactions between water and soil are most important (Santschi & Schwehr, 2004). Beside physical and chemical factors, biological processes especially promoted by microorganism influence the cycling of iodine. Microorganisms are involved in environmental processes as primary producers and also as consumers and decomposers. They have bioremedial and biotransformable potential and in this way affect the mobility of elements. Oxidation and reduction mechanisms contribute to transformations between soluble and insoluble forms. Experiments with ^{125}I tracer showed the importance of microbial participation in iodine accumulation – sorption and desorption processes – in soil. Muramatsu et al. (1996) observed desorption of iodine from flooded soil during cultivation of rice plants. Microorganisms created reducing conditions in the flooded soil and iodine once adsorbed on the soils was desorbed (Muramatsu et al., 1996). Amachi et al. (2001) reported a wide variety of terrestrial and marine bacteria that are capable to produce CH_3I under oligotrophic conditions. Aerobic bacteria showed significant production of CH_3I , whereas anaerobic did not produce it. The methylation of iodide was catalysed enzymatically with S-adenosyl-L-methionine as the methyl donor.

The binding of iodine by organic matter and/or iron and aluminium oxides has the potential to modify the transport, bioavailability and transfer of iodine isotopes to man (Santschi & Schwehr, 2004). Because of the same chemical properties ^{129}I and ^{127}I should behave similar in environmental processes. Major pathways are the volatilization of organic iodine compounds into the atmosphere, accumulation of iodine in living organisms, oxidation and reduction of inorganic iodine species, and sorption of iodine by soils and sediments. These processes are influenced or even controlled by microbial activities (Amachi, 2008).

^{129}I is gradually released in trace quantities into the atmosphere and aquatic environment from reprocessing plants. It is then physically transported in the air or water media under the influence of chemical and biological processes. Newly introduced ^{129}I from NFRP is in volatile form and as such more mobile compared to ^{127}I . By taking this aspect into account one cannot be sure that biogeochemical behaviour of ^{129}I and ^{127}I is the same. Even more, Santschi & Schwehr (2004) discussed that biogeochemical behaviour of iodine and its isotopes appears to be different in North American and European waters.

4. Measurement of ^{129}I

^{129}I decays by emitting beta particles ($E_{\beta\text{max}} = 154.4 \text{ keV}$), gamma rays ($E_{\gamma} = 39.6 \text{ keV}$) and X-rays (29–30 keV) to stable ^{129}Xe (Tendow, 1996). Therefore it can be measured by gamma and X-ray spectrometry and by beta counting using liquid scintillation counters (LSC).

Another method for determination of ^{129}I is neutron activation analysis (NAA) that is based on neutron activation of $^{129}\text{I}(n, \gamma)^{130}\text{I}$, which is measured by gamma spectrometry ($E_{\gamma} = 536 \text{ keV}$ (99 %)). In recent year's mass spectrometry – such as accelerator mass spectrometry (AMS) and inductively coupled plasma mass spectrometry (ICP-MS) are also used.

For determination of ^{129}I levels in environmental samples only two analytical methods are available, radiochemical neutron activation analysis (RNAA) and AMS. The main advantage of the AMS is the detection limit that is close to 10^{-14} expressed as $^{129}\text{I}/^{127}\text{I}$ ratio. RNAA can only measure ^{129}I at elevated levels – nuclear era. AMS enables measurement of ^{129}I in all environmental samples, also the natural, pre-nuclear levels, and the needed amount of sample is 10-100 times smaller than in the case of RNAA. Detection limits for ^{129}I using different analytical methods are compared in Table 6.

Analytical method/Sample	Detection limit		Reference
	$\text{g g}^{-1} (10^{-12})$	$^{129}\text{I}/^{127}\text{I} (10^{-12})$	
<i>γ-X spectrometry</i>			
seaweed (400 g)	300	not given	Lefevre et al., 2003
<i>LSC</i>			
radioactive waste (coolant, 1 L)	23	not given	Gudelis et al., 2006
<i>ICP-MS</i>			
Aqueous solution	100	1000000	Muramatsu et al., 2008
Aqueous solution	0.8	not given	Izmer et al., 2003
Aqueous solution (groundwater)	5	not given	Brown et al., 2007
Sediment	30	not given	Izmer et al., 2003
Sediment	0.4	not given	Izmer et al., 2004
<i>RNAA</i>			
soil (100 g)	0.05	5000	Osterc et al., 2007
soil (100 g)	0.015	10000	Muramatsu & Yoshida, 1995
soil (80 g)	0.27	not given	Michel et al., 2005
soil	0.13	410	Szidat et al., 2000b
<i>AMS</i>			
commercial AgI	not given	0.44	Suzuki et al., 2006
blank sample	not given	0.50	Gomez-Guzman et al., 2011
blank sample	not given	0.17	Muramatsu et al., 2008
soil (1 g)	0.0015	40	
soil (80 g)	0.00015	5	Michel et al., 2005
Woodward Iodine*	not given	0.023	Reithmeier et al., 2005
Woodward Iodine	not given	0.04	Buraglio et al., 2001
oil and gas hydrates	not given	0.20	Alfimov & Synal, 2010
soil	0.000023	0.75	Szidat et al., 2000b

*Woodward Iodine is elemental iodine mined by Woodward Iodine Corp. in Oklahoma for which the lowest ratio is reported.

Table 6. Limits of detection for ^{129}I in various samples using different analytical methods

4.1 Direct gamma and X-ray spectrometry

Direct gamma-X spectrometry ($E_{\gamma} = 39.6$ keV; X-rays, 29–30 keV) is a non-destructive technique that is rapid and can be applied to different matrices. It is used for monitoring of environmental samples collected in vicinity of NFRP such as thyroid, urine, seaweed, and for nuclear waste by using high purity Ge or plenary Si detector (Suarez et al., 1996; Bouisset et al., 1999; Frechou et al., 2001; Lefevre et al., 2003; Frechou & Calmet, 2003; Barker et al., 2005). To lower the detection limits normally big samples (50–500 g) are used, which induces considerable attenuation at low energies. The attenuation depends on the matrix composition of the sample and geometric parameters of the container. Therefore the mass energy-attenuation coefficient (self-absorption correction) at a given energy must be measured for all sample matrices with respect to that of the standard source. Experimentally obtained self-absorption correction factors are used to obtain accurate results (Bouisset et al., 1999; Lefevre et al., 2003, Barker et al., 2005). To quantify self-absorption correction factors ^{210}Pb (46.5 keV) and ^{241}Am (59.6 keV), with gamma lines close to ^{129}I are used. Detection limits as low as 2 Bq kg⁻¹ dry mass can be reached for *Fucus sp.* samples (Bouisset et al., 1999).

Chemical separation of ^{129}I from the sample matrix and interfering radionuclides – destructive method – improves the detection limit when using direct gamma-X spectrometry (Suarez et al., 1996).

By using direct gamma-X spectrometry ^{129}I was determined in seaweed sample FC-98 Seaweed, which was prepared by Frechou et al. (2001), by using direct gamma -X spectrometry (Osterc & Stibilj, 2008).

4.2 Liquid Scintillation Counting (LSC)

Liquid scintillation counting is based on emissions of beta particles from radionuclides – beta decay ($E_{\beta\text{max}} = 154.4$ keV). ^{129}I has to be separated from the sample matrix and other radionuclides and dissolved or suspended in a scintillation cocktail containing an organic solvent and a scintillator. Beta particles emitted from the sample transfer energy to the solvent molecules, which in turn transfer their energy to the scintillator which relaxes by emitting light - photons. In a liquid scintillation counter each beta emission (ideally) results in a pulse of light, which is amplified in a photomultiplier and detected.

Recently extraction chromatographic resins for the separation and determination of ^{36}Cl and ^{129}I have been developed. First results show a promising potential to use the resins within the context of the monitoring of nuclear installations – during operation and especially during decommissioning (Zulauf et al., 2010).

4.3 Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

ICP-MS has been used to determine ^{129}I in contaminated environmental samples with high level ^{129}I content such as sediments, groundwater samples, soil and seaweed (Izmer et al., 2003; Izmer et al., 2004; Becker, 2005; Brown et al., 2007; Li et al., 2009). The lowest detection limit of the method reported as $^{129}\text{I}/^{127}\text{I}$ isotopic ratio is 10⁻⁷.

The method is based on iodine separation and injection to the machine as solution or gaseous iodine, I₂. Iodine is decomposed into iodine atom and ionized to positive iodine ion at a temperature ~6000–8000 K. It is then extracted from the plasma into a high vacuum of the mass spectrometer via an interface. The extracted ions are separated by mass filters of

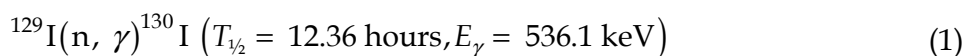
either quadropole type time-of-flight or combination of magnetic and electrostatic sector and measured by an ion decetor (Hou et al., 2009).

Difficulties encountered when determining ^{129}I with ICP-MS are low ^{129}I quantities present with high ^{127}I concentrations, isobaric and molecular ions interferences ($^{129}\text{Xe}^+$, $^{127}\text{IH}_2^+$), memory effects and tailing of ^{127}I . To improve $^{129}\text{I}/^{127}\text{I}$ determination it was found that introduction of helium gas into collision cell reduces peak tail of a high-abundant isotope, ^{127}I by up to three orders of magnitude. Detection limits have been improved by applying oxygen as collision gas for selective reduction of ^{129}Xe (Izmer et al., 2003, Hou et al., 2009).

4.4 Neutron Activation Analysis (NAA)

NAA enables determination of ^{129}I in environmental samples at 10^{-10} $^{129}\text{I}/^{127}\text{I}$ isotopic ratios. The concentration levels of ^{129}I in environmental samples are very low and chemical separation/pre-concentration procedures have to be developed which can be used for a wide variety of matrices.

Neutron activation analysis is based on induction of ^{129}I with thermal neutrons – irradiation in a nuclear reactor via following nuclear reaction:



^{129}I is determined by measuring of ^{130}I activity on a high purity Ge detector. Interfering nuclear reactions induced during irradiation of sample from other nuclides resulting in ^{130}I production can influence the correct determination of ^{129}I . These undesired nuclides are ^{235}U , ^{128}Te and ^{133}Cs and nuclear reactions: $^{235}\text{U}(n, f)^{129}\text{I}(n, \gamma)^{130}\text{I}$, $^{235}\text{U}(n, f)^{130}\text{I}$, $^{128}\text{Te}(n, \gamma)^{129\text{m}}\text{Te}(\beta^-)^{129}\text{I}(n, \gamma)^{130}\text{I}$ and $^{133}\text{Cs}(n, \alpha)^{130}\text{I}$ (Hou et al., 1999). They have to be removed from the sample before irradiation to avoid nuclear interferences.

During irradiation radioactivity in sample is produced mainly due to the radioisotopes $^{23}\text{Na}(n, \gamma)^{24}\text{Na}$ ($T_{1/2} = 14.96$ hours), $^{41}\text{K}(n, \gamma)^{42}\text{K}$ ($T_{1/2} = 12.36$ hours) and $^{81}\text{Br}(n, \gamma)^{82}\text{Br}$ ($T_{1/2} = 35.30$ hours) present in sample, which renders the direct measurement of ^{130}I after irradiation and radiochemical separation of induced ^{130}I after irradiation is necessary. Solvent extraction with CCl_4 or CHCl_3 are normally used to extract iodine (Osterc & Stibilj, 2005; Osterc et al., 2007).

In first step pre-concentration of iodine from large amounts of sample is performed. Solid samples, such as soil, sediment, vegetation, biological samples can be decomposed by alkaline fusion (Hou et al., 1999, Osterc et al., 2007). The sample is mixed with potassium hydroxide/alkali solution and then gradually heated to 600°C . Iodine is leached from the decomposed sample with hot water, isolated with solvent extraction and precipitated as PdI_2 or MgI_2 or trapped on activated charcoal (Fig. 2) (Hou et al., 1999, Osterc et al., 2007). Another method to separate iodine from solid samples is combustion at high temperature, $\sim 1100^\circ\text{C}$ (Muramatsu & Yoshida, 1995). Released iodine is trapped in an alkaline solution or adsorbed on activated charcoal.

The pre-concentrated iodine is than irradiated for up to 12 hours simultaneously with a $^{129}\text{I}/^{127}\text{I}$ standard. After radiochemical separation the ^{130}I induced from ^{129}I (see nuclear reaction 1) is counted on a high purity Ge detector and compared to standard of known activity and corrected for chemical yield (Osterc et al., 2007).

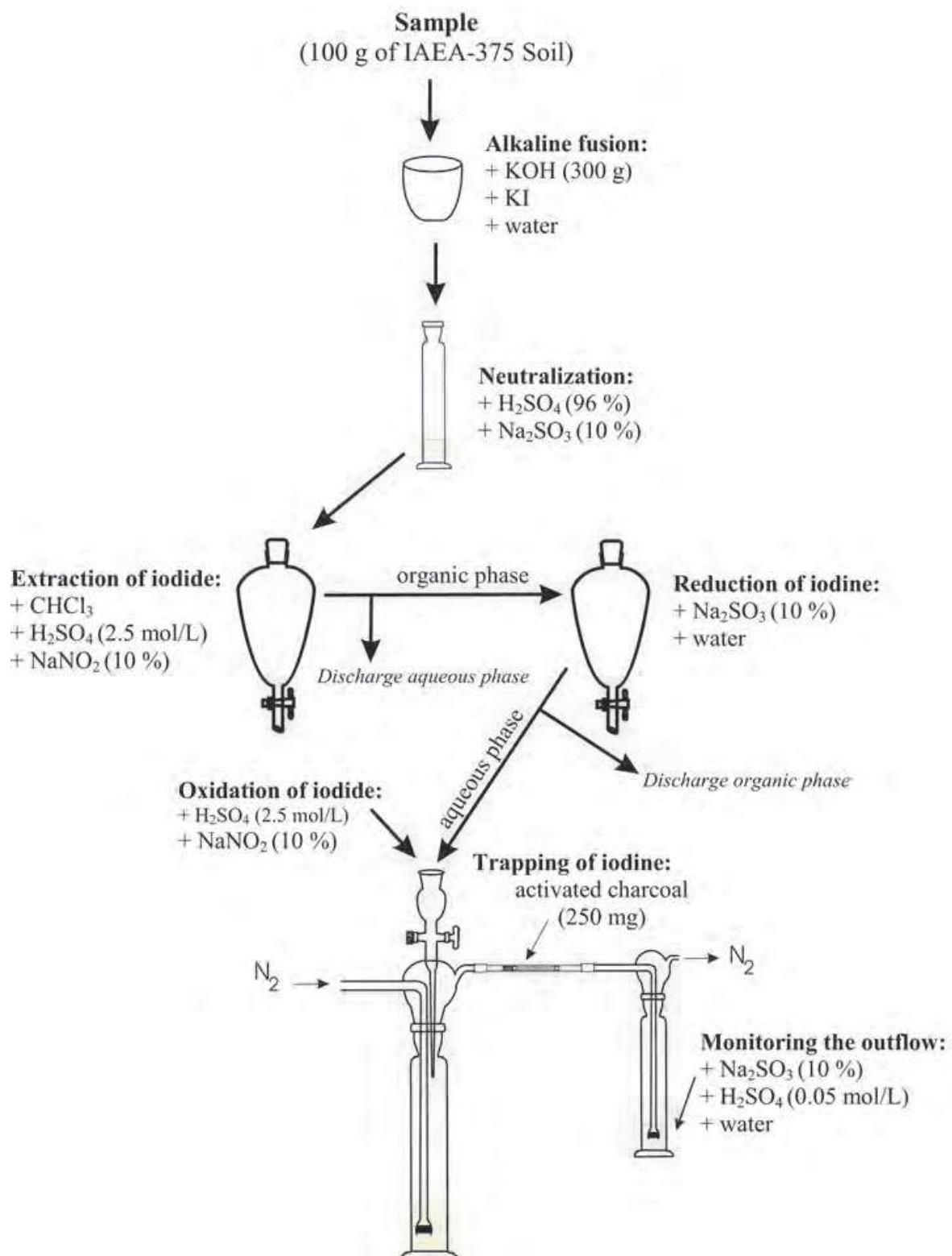


Fig. 2. The scheme for pre-concentration of iodine from solid samples (Osterc et al., 2007)

For liquid samples, such as milk, urine and water samples anion exchange method using anion exchange resins can be applied. Adsorbed iodide is eluted and isolated from the eluate with solvent extraction and precipitated as PdI₂ or MgI₂ (Parry et al., 1995; Hou et al., 2001; Hou et al., 2003a).

4.5 Accelerator Mass Spectrometry (AMS)

An AMS facility is set up off injector and analyser linked with a tandem accelerator. The detector is either a combination of time-of-flight and silicon charged particle detector or gas ionization energy detector. Iodine has to be separated from the sample with same techniques as used for NAA, such as pyrohydrolysis at 1000 °C, and prepared as AgI targets (Muramatsu et al., 2008). Negative iodine ions are produced from AgI targets by Cs sputter ion source and injected into the tandem accelerator. The formed $^{129}\text{I}^-$ and $^{127}\text{I}^-$ ions are accelerated to positive high-voltage terminal converting negative ions to I^{3+} , I^{5+} or I^{7+} . The positively charged ions pass through a magnetic analyser where ions of ^{129}I and ^{127}I based on charge state and energy are selected and directed to a detector. AMS measures the $^{129}\text{I}/^{127}\text{I}$ isotopic ratio and the ^{129}I absolute concentration is calculated by the ^{127}I content determined in the sample and the chemical yield for separation of iodine from sample – preparation of AgI targets (Hou et al., 2009).

AMS is the only technique that enables measurement of pre-nuclear age samples and samples with low ^{129}I content, below 10^{-10} $^{129}\text{I}/^{127}\text{I}$ isotopic ratio (Moran et al., 1998; Fehn et al., 2000a; Buraglio et al., 2001; Alfimov et al., 2004; Santschi & Schwehr, 2004; Snyder & Fehn, 2004; Michel et al., 2005; Fehn et al., 2007; Hou et al., 2007; Keogh, et al., 2007; Muramatsu et al., 2008; Gomez-Guzman et al., 2011). Instrumental background of 10^{-14} $^{129}\text{I}/^{127}\text{I}$ has been obtained (Buraglio et al., 2000). But the detection limit depends on the chemical separation before measurement and especially on addition of iodine carrier. When carrier and chemical processing are included the typical reported blank $^{129}\text{I}/^{127}\text{I}$ isotopic ratio is $1 \cdot 10^{-13}$ (Buraglio et al., 2000). For environmental samples with a very low $^{129}\text{I}/^{127}\text{I}$ isotopic ratio Hou et al. (2010) reported a method for preparation of carrier free AgI targets based on co-precipitation of AgI with AgCl to exclude the influence of interferences from ^{129}I and ^{127}I in the carrier. They calculated a detection limit of 10^5 atoms, which corresponds to $2 \cdot 10^{-16}$ g of ^{129}I .

4.6 Quality assurance of ^{129}I analyses

To be able to determine ^{129}I by RNAA in environmental samples from nuclear era pre-concentration of iodine from large amounts of sample (up to 150 g) is needed. In this pre-concentration step contamination of sample with ^{129}I is possible. It is important to make a blank control when establishing a new method and verify the method by reference materials to evaluate possible contamination during the entire analytical process; including pre-concentration, irradiation, radiochemical separation and gamma activity measurement.

Also analysis of ^{129}I by AMS requires intensive and continuous control – control charts of the analytical blank and verification of accuracy by analysis of reference materials, which has to be continued periodically also during routine operation (Szidat et al., 2000a). Influence of sample mass – AgI targets on accuracy of ^{129}I determination was studied by Lu et al. (2007). They found that samples with masses above 0.3 mg did not show an influence on accuracy – ion current of the sample was constant, but it fell strongly for samples with masses below 0.3 mg. Samples with masses below 0.1 mg did not produce sustainable currents for ^{129}I determination. Presence of 5000 ^{129}I atoms or 50 μg in the target is sufficient for a successful ^{129}I determination. To validate and or evaluate an analytical method, to run a laboratory inter-comparison, to check accuracy of analytical method, and ensure globally comparable and traceable results to stated references, as the SI units, certified reference materials are needed. Environmental samples represent a huge variety of different combinations of substances to be analysed and the matrices in which they are embedded. This countless combinations of substances –

elements, radionuclides, contaminants – and matrices means that certified reference materials always lack.

The only reference material with a recommended value for ^{129}I available on the market was the reference material IAEA-375 Soil – Radionuclides and Trace Elements in Soil. Top soil to a depth of 20 cm was obtained from the “Staryi Viskov” collective farm in Novozybkov, Brjansk, Russia in July 1990. Unfortunately this reference material is now out of stock.

Only informative and not certified values for ^{129}I , determined in one laboratory, are reported for NIST SRM 4357 – Ocean Sediment Environmental Radioactivity Standard, which is a blend of ocean sediments collected off the coast of Sellafield, UK, and in the Chesapeake Bay, USA, and NIST SRM 4359 – Seaweed Radionuclide Standard, which is a blend of seaweed collected off the coast of Ireland and the White Sea.

Recently a new reference material, with a certified value for ^{129}I , IAEA-418: I-129 in Mediterranean Sea Water was characterised in an interlaboratory comparison exercise. The used method was AMS (accelerator mass spectrometry).

Another new reference material for radionuclides in the mussel *Mytilus galloprovincialis* from Mediterranean Sea, IAEA-437 was characterised. They reported for the mussel sample collected in 2003 at Anse de Carteau, Port Saint Louis du Rhône, France an informative average massic activity of $0.8 \pm 0.1 \text{ mBq kg}^{-1}$ dry mass (Pham et al., 2010).

5. Applications of I-129 as an environmental tracer

Use of ^{129}I as an intrinsic tracer for natural iodine kinetics was discussed as early as 1962 (Edwards, 1962). Already at that time two reprocessing plants, one for military purposes in Marcoule, France (from 1958) and one for nuclear fuel in Thurso, United Kingdom (from 1958) existed.

To be able to use ^{129}I as an environmental tracer certain conditions have to be met. These are: (1) ^{129}I must trace a single environmental process with a defined time scale; (2) ^{129}I must be equilibrated with ^{127}I ; (3) The predominant chemical species of ^{129}I and their geochemical properties must be known (Santschi & Schwehr, 2004); (4) Conservative behaviour, meaning relatively constant concentration in a reservoir over time, is desirable. The natural $^{129}\text{I}/^{127}\text{I}$ ratio has been strongly shifted by continuous additions from anthropogenic sources, which still persists. To trace existing and future global changes in inventories of anthropogenic ^{129}I continuous monitoring and revised budget calculation are indispensable (Aldahan et al. 2007a). Recently also a prediction model system to better understand the dispersion of ^{129}I from point sources (Sellafield and La Hague) to the northern North Atlantic Ocean has been developed (Orre et al. 2010).

United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000) identifies as globally dispersed radionuclides ^3H , ^{14}C and ^{129}I . Because of its very long half live is ^{129}I one of the most important radionuclides in long-term radiological assessment of its discharges from nuclear fuel reprocessing plants. ^{129}I is present in the environment in low quantities (in traces) and its increase in a particular compartment of the ecosystem can be instantly recognized.

5.1 ^{129}I as an oceanographic tracer

Transport, circulation and exchange of water masses in the Northeast Atlantic and Arctic Oceans has long been studied by using radionuclides such as ^{137}Cs , ^{134}Cs , ^{90}Sr , ^{125}Sb and $^{99\text{m}}\text{Tc}$ originating from reprocessing of spent nuclear fuel. In recent years ^{129}I became

interesting as an oceanographic tracer, because the discharges from NFRP in La Hague and Sellafield increased since 1990 and highly sensitive analytical method, AMS, developed for analysis (Hou, 2004).

Concentrations and species of ^{129}I and $^{129}\text{I}/^{127}\text{I}$ isotopic ratio were determined in many environmental and biological samples from marine environment, especially in areas influenced by NFRP. Results for Northeast Atlantic, Arctic and Baltic Seas indicate a strong influence of liquid discharges from NFRP in La Hague and Sellafield. Hou et al. (2000a) determined ^{129}I concentrations in archived time series seaweed *Fucus vesiculosus* samples from Danish, Norwegian and Northwest Greenland coast collected in a period from 1980 to 1997 (Table 3). They used the $^{129}\text{I}/^{99}\text{Tc}$ ratio to estimate the origin of and transit times of ^{129}I . Transit times were estimated to be 1–2 years from La Hague, 3–4 from Sellafield, to Denmark (Klint) and Norway (Utsira), and 9–14 years from La Hague, 11–16 from Sellafield, to NW Greenland.

Iodine exists in seawater mainly as dissolved iodate and iodide, and a small amount of organic iodine (Wong, 1991). Chemical speciation of ^{129}I can be used to investigate the transport, dispersion, and circulation of the water masses – especially at the boundary of two or more sources. (Hou et al., 2001).

5.2 ^{129}I as a geochemical tracer

^{129}I was used in geochemical studies as a tracer for determining ages and migration of brines (Muramatsu et al., 2001, Snyder et al., 2003a, Fehn et al., 2007). Isolated system contain lower or close to estimated pre-nuclear $^{129}\text{I}/^{127}\text{I}$ ratio, $1.5 \cdot 10^{-12}$. For correct interpretation of results – age calculation based on ^{129}I one must consider the effect of possible fissiogenic production and initial concentration on isotopic ratios. The estimated pre-nuclear ratio can be disturbed along continental margins with lower isotopic ratios likely caused by releases of methane-rich fluids with high stable iodine concentrations derived from old organic sources, where ^{129}I already partly decayed. The isotopic ratio of the open ocean is not disturbed, justifying the use of estimated pre-nuclear ratio (Fehn et al., 2007).

5.3 ^{129}I in precipitation

Atmospheric releases of ^{129}I from European and Hanford NFRP were much higher than from nuclear weapons tests and Chernobyl accident together (Table 1). Measurement of ^{129}I in atmosphere and precipitation can be used to investigate the transport pathways of ^{129}I from point sources, such as NFRP. But it is important to be aware that ^{129}I levels in atmosphere and precipitation can originate either directly from atmospheric releases from NFRP, and from volatilization from seawater and terrestrial environment. To study transport pathways of ^{129}I all of this aspects have to be considered and obtained results for atmospheric and precipitation samples compared to reported releases from NFRP in particular timescale. Many precipitation and atmospheric samples have to be measured continuously to establish a pattern or trend.

5.4 ^{129}I for reconstruction of ^{131}I dose

The same chemical and physical properties of isotopes of particular element enable to use ^{129}I as a tool for the reconstruction of ^{131}I doses after a nuclear accident. This was done after the nuclear accident in Chernobyl. Levels of ^{129}I were determined in soils and from the measured $^{129}\text{I}/^{131}\text{I}$ ratio, 12–19 (Kutschera et al., 1988; Mironov et al., 2002), the long-lived

^{129}I can be used to reconstruct ^{131}I dose to thyroids. This method is limited only to areas that were relatively strong contaminated by fallout from Chernobyl like areas in Ukraine and Belarus (Michel et al., 2005; Straume et al., 2006).

6. Radiological hazard of ^{129}I for man

Transport pathways of iodine to human are ingestion and inhalation. Iodine present in food is adsorbed into blood in small intestine – inhaled iodine from the air is also transferred into blood. More than 80 % of iodine absorbed into the blood is concentrated in the thyroid gland, which is therefore the target organ of iodine – also radioactive ^{129}I . Due to low beta and gamma energy of ^{129}I and long half-life the radiation toxicity of ^{129}I is mainly related to long term and low dose internal exposure of the thyroid to the beta radiation of ^{129}I . An average iodine content in human thyroid is 10–15 mg. ^{129}I and ^{127}I are taken up by thyroid indiscriminately. The highest reported $^{129}\text{I}/^{127}\text{I}$ ratio was 10^{-4} in close vicinity of NFRP, which corresponds to 10^{-6} g or 6.64 Bq at 10 mg stable iodine content in thyroid. The corresponding annual radiation dose to thyroid would be 0.1 mSv year $^{-1}$, which is 2.5 times higher than the dose regulation limit of 0.04 mSv year $^{-1}$ set by the U.S. NRC for combined beta and photon emitting radionuclide to the whole body or any organ (Hou et al., 2009). An annual thyroid equivalent dose of 1 mSv, which is comparable to the level of natural back-ground radiation, would only be reached by ratios exceeding $1.5 \cdot 10^{-3}$ (Michel, 1999). Current concentrations of ^{129}I in the environment do not represent any radiological hazard for man, even in the vicinity of nuclear fuel reprocessing plants. But to assess environmental impact and potential risk and consequences during long-term exposition information on the distribution and radionuclide species, speciation analysis, influencing the mobility, biological uptake and accumulation of radionuclides is needed (Salbu, 2007). Speciation analysis provides crucial information for evaluation of radionuclide transport mechanism in the environment and to the human body and accurate risk assessments (Hou et al., 2009).

7. Conclusion

Anthropogenic ^{129}I considerable enriched pre-nuclear environmental levels. Presently the main sources of ^{129}I in the environment are nuclear fuel reprocessing plants (NFRP). Global distribution of ^{129}I is not uniform – concentrations are elevated near NFRP – but anthropogenic ^{129}I was detected in remote areas such as Antarctic.

Before the onset of nuclear age ^{129}I and ^{127}I were in equilibrium. Analysis of pre-nuclear material and deep layer of marine sediment gave the best estimated value for natural $^{129}\text{I}/^{127}\text{I}$ ratio in surface reservoirs to be $(1.5 \pm 0.15) \cdot 10^{-12}$.

In transport and exchange of ^{129}I among different compartments marine and soil ecosystems influenced by present biota – microorganisms play major role. Biogeochemical cycling of iodine is influenced by its strong association with organic material – ocean is the main reservoir of mobile iodine, where it is rapidly exchanged between biota, hydrosphere and atmosphere.

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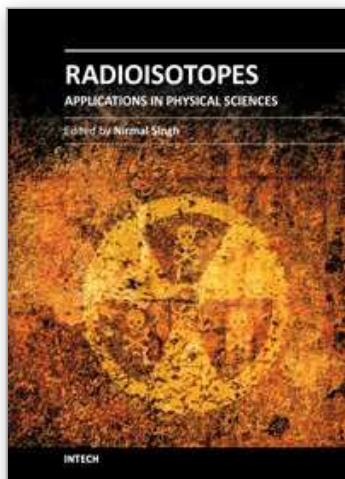
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The book Radioisotopes - Applications in Physical Sciences is divided into three sections namely: Radioisotopes and Some Physical Aspects, Radioisotopes in Environment and Radioisotopes in Power System Space Applications. Section I contains nine chapters on radioisotopes and production and their various applications in some physical and chemical processes. In Section II, ten chapters on the applications of radioisotopes in environment have been added. The interesting articles related to soil, water, environmental dosimetry/tracer and composition analyzer etc. are worth reading. Section III has three chapters on the use of radioisotopes in power systems which generate electrical power by converting heat released from the nuclear decay of radioactive isotopes. The system has to be flown in space for space exploration and radioisotopes can be a good alternative for heat-to-electrical energy conversion. The reader will very much benefit from the chapters presented in this section.

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University Campus STeP Ri
Slavka Krautzeka 83/A
51000 Rijeka, Croatia
Phone: +385 (51) 770 447
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Unit 405, Office Block, Hotel Equatorial Shanghai
No.65, Yan An Road (West), Shanghai, 200040, China
中国上海市延安西路65号上海国际贵都大饭店办公楼405单元
Phone: +86-21-62489820
Fax: +86-21-62489821

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