Long-Lived Radionuclides in the Environment: The Case of Iodine-129

R. Michel, Th. Ernst, D. Jakob, K. Klipsch, S. Szidat*, H.-A. Synal**, Ch. Schnabel***,****

Center for Radiation Protection and Radioecology (ZSR), University Hanover, Am Kleinen Felde 30, D-30167 Hannover, Germany
* present address: Department of Chemistry and Biochemistry, University Bern; Switzerland
** Paul Scherrer Institute, c/o Institute for Particle Physics, ETH Hoenggerberg, Zurich, Switzerland
*** Institute for Particle Physics, ETH Hoenggerberg, Zurich, Switzerland
**** Scottish Universities Environmental Research Centre, Scottish Enterprise Technology Park, East Kilbride G75 0QF, Great Brittain

Abstract: Results are presented from an ongoing project to study the status and behavior of [129]I and [127]I in the environment. [129]I and [127]I abundances were investigated in precipitation, surface and ground waters from Lower Saxony, Germany, and in soil samples from various European locations. From the analysis of [129]I in rain, [129]I annual deposition densities were determined for the time period from 1997- 1999. We conclude that [129]I deposition rates in Switzerland and Germany increased by three orders of magnitude since 1950 and changed just little after 1987. From the analysis of soils, [129]I deposition densities at various places of Europe were determined. Thereby, the [129]I natural equilibrium deposition density as well as that of the fall-out from atmospheric weapon tests was estimated. Elevated [129]I abundances in Ukrainian soils contaminated by Chernobyl fall-out provide a basis for retrospective dosimetry of the radiation exposure due to [131]I. Soil profiles from Germany exhibit the influence of ongoing emissions from European reprocessing plants and demonstrate the complexity of iodine migration. Biospheric [129]I/[127]I ratios in Germany are an order of magnitude lower than in precipitation and . Because of the disequilibrium of iodine isotopes in the different compartments further detailed investigations of the pathways of [129]I through the environment to man are considered necessary.

1. INTRODUCTION

The long-lived radionuclide [129]I ($T_{1/2}$ = 15.7 Ma, $E_{\beta,max}$ =0.2 MeV, E_γ = 39.58 keV, I_γ = 0.0752) is produced in nature by cosmic-ray induced spallation of xenon in the atmosphere and by spontaneous fission in the geosphere. The total natural inventory of [129]I in the lithosphere was estimated to be 50 000 kg (327 TBq) and of this only 263 kg (1.7 TBq) being available as "free" inventory of atmosphere, hydrosphere and biosphere and mixing with the stable [127]I; see [1] for details. The natural abundances of [129]I have been globally and sustainably changed by fissionogenic [129]I released by man into the environment. Estimates of the [129]I releases from explosions of nuclear weapons are between 43 kg (0.28 TBq) and 150 kg (0.98 TBq) [2-5]. In the Chernobyl accident less than 2 kg (0.013 TBq) [129]I was emitted [1]. The vast amount of man-made [129]I in the environment is due to releases from reprocessing plants. Up to the year 2000, the European reprocessing plants emitted about 3500 kg (23 TBq) [129]I, of this 70 % being released by the La Hague reprocessing plant [6-11]. The emissions were mainly liquid discharges, 97 % of the [129]I at La Hague and about 85 % at Sellafield. Large amounts of man-made [129]I are still stored in spent nuclear fuel. In 1990 this [129]I was estimated to amount to 5660 kg (37 TBq) [12]. Information about the releases from worldwide military reprocessing plants and about plants in the former USSR and other eastern countries are lacking.

Man-made [129]I has globally changed the natural [129]I/[127]I ratios in the oceanic mixing layer by two orders of magnitude from about 1.5×10^{-12} [13,14] to about 10^{-10} [12] remote from emittents with orders of magnitude higher ratios close to the release points [12]. Though these changes are of radiological relevance only in the closest proximity of reprocessing plants, they should be carefully monitored and the potential of [129]I as a man-made tracer for environmental processes should be

exploited. This requires a detailed understanding of the pre-nuclear baseline values, of the anthropogenic changes, of the radioecology of [129]I, and of the environmental chemistry of iodine. However, due to analytical and methodological problems the radioecology of [129]I is still insufficiently known [1]. We therefore started systematic investigations to establish reliable analytical protocols for [129]I and [127]I analyses in various environmental materials [1,15-18], to improve our knowledge about the environmental abundances and the behavior of [129]I, and to provide the baseline data for some tracer applications, in particular with respect to retrospective dosimetry [19,20].

2. Materials and Methods

Since 1997, precipitation, surface waters and ground water samples from Lower Saxony, Germany, were collected at various stations of the Deposition Measurement System. Moreover, surface and shallow ground water samples were taken at stations of the Information System of Surveillance of Environmental Radioactivity (IMIS). In order to establish a spatial separation, Lower Saxony was divided into four regions with sampling sites of every sample type each. The four regions were chosen to represent the close proximity to the North Sea (region I), northern German lowlands distant from the North Sea (region II), the fringe of the Harz mountains (region III) and the area of the Elbe river in Lower Saxony in the vicinity of Gorleben (region IV). In addition to open field precipitation sampled in each region, through-fall precipitation was collected in two regions; through-fall here means rain that falls down on forests and interacts with the canopy of the trees on its way down. Samples of North Sea water were taken singularly near the German shore in 1999 and 2000 at Spieka-Neufeld and Amrum. [129]I and [127]I were analyzed by AMS and ICP-MS, respectively [18].

Soil samples were obtained from different sources. 68 soil profiles down to a depth of 40 cm were taken between 1995 and 1997 at Moscow and in Northern Ukraine, of which 44 profiles were analyzed so far for [129]I and [127]I using RNAA and AMS. A pre-nuclear soil profile from 1939 was obtained from the Timiryazev Agricultural Academy, Moscow, courtesy of Prof. Dr. A. Fokin. In 1999, seven well characterized soil profiles down to a maximum depth of 250 cm from seven locations in Lower Saxony were taken and analyzed by IC and AMS for [127]I and [129]I, respectively [21].

The development with time of [129]I/[127]I isotopic ratios in human and animal thyroid glands sampled between 1987 and 1993 was systematically investigated by Handl and coworkers; [22] and references therein. Here we report new data obtained by RNAA on human thyroid glands sampled between 1999 and 2001 in Lower Saxony/Germany.

Detailed descriptions of the sampling procedures, sample preparations, analytical techniques (AMS, RNAA, ICP-MS and IC), and quality control measures are given elsewhere [1,15-18].

3. ATMOSPHERIC FALL-OUT OF [129]I IN GERMANY AND SWITZERLAND

The geometric means and standard deviations of [129]I, [127]I and of [129]I/[127]I ratios in North Sea water, precipitation, surface and ground waters from Lower Saxony, Germany, are given in table 1. The detailed results are presented elsewhere [17,18]. We will focus first on the discussion of the [129]I/[127]I ratios. According to model calculations, e.g. [23], iodine pathways through the environment are recognizable via these [129]I/[127]I ratios. The North Sea and the North Atlantic act as buffer reservoirs for both, stable iodine and [129]I, the latter being mainly emitted by the European reprocessing plants Sellafield and La Hague. The subsequent transfer through marine and land atmospheres to precipitation takes place with only a short time scale of about two weeks [23]. In the atmosphere, mixing with iodine from other natural sources occurs and also a direct influence of atmospheric releases from reprocessing plants to [129]I in precipitation is possible. After iodine is deposited on the ground by wet or dry precipitation, the iodine isotopes are slowly transported into surface and ground waters. These compartments have distinctly lower [129]I/[127]I ratios, which makes it possible to estimate residence times of iodine in catchments via the time dependence of [129]I abundances [24]. As iodine is biophilic, these residence times may provide evidence regarding the biological condition of a catchment when compared to tritium residence times.

Table 1: [129]I and [127]I activity concentrations and [129]I/[127]I isotopic ratios in precipitation, surface and ground waters from Lower Saxony/Germany (1997-1999) and in North Sea water (1999 -2000) [17,18,25]. The data are given as geometric means with standard deviations. For a description of the different regions see section2.

sample	region	n	[129]I in µBq kg⁻¹	[127]I in µg kg⁻¹	[129]I/[127]I in 10^{-10}
North Sea water	I	5	$495 \times 1{,}73^{\pm1}$	$44{,}9 \times 1{,}82^{\pm1}$	$16500 \times 1{,}24^{\pm1}$
precipitation open field	I	10	$17 \times 1{,}34^{\pm1}$	$3{,}1 \times 1{,}39^{\pm1}$	$8250 \times 1{,}20^{\pm1}$
	II	9	$6{,}07 \times 1{,}41^{\pm1}$	$1{,}48 \times 1{,}31^{\pm1}$	$6190 \times 1{,}34^{\pm1}$
	III	9	$4{,}09 \times 1{,}87^{\pm1}$	$1{,}33 \times 2{,}05^{\pm1}$	$4650 \times 1{,}82^{\pm1}$
	IV	9	$3{,}57 \times 1{,}61^{\pm1}$	$1{,}31 \times 1{,}45^{\pm1}$	$3740 \times 1{,}59^{\pm1}$
precipitation through-fall	II	9	$24{,}1 \times 1{,}40^{\pm1}$	$6{,}92 \times 1{,}31^{\pm1}$	$5260 \times 1{,}13^{\pm1}$
	III	9	$14{,}2 \times 1{,}33^{\pm1}$	$5{,}08 \times 1{,}23^{\pm1}$	$4220 \times 1{,}29^{\pm1}$
surface water	I	12	$7{,}2 \times 2{,}25^{\pm1}$	$23{,}6 \times 1{,}48^{\pm1}$	$460 \times 2{,}07^{\pm1}$
	II	12	$0{,}94 \times 1{,}67^{\pm1}$	$6{,}52 \times 1{,}25^{\pm1}$	$219 \times 1{,}61^{\pm1}$
	III	11	$0{,}24 \times 1{,}58^{\pm1}$	$5{,}74 \times 1{,}43^{\pm1}$	$62{,}8 \times 1{,}68^{\pm1}$
	IV	12	$0{,}67 \times 2{,}00^{\pm1}$	$8{,}56 \times 1{,}61^{\pm1}$	$118 \times 2{,}04^{\pm1}$
groundwater	I	3	$0{,}283 \times 1{,}39^{\pm1}$	$4{,}24 \times 1{,}30^{\pm1}$	$99{,}2 \times 1{,}51^{\pm1}$
	II	4	$0{,}56 \times 1{,}23^{\pm1}$	$3{,}9 \times 1{,}26^{\pm1}$	$222 \times 1{,}56^{\pm1}$
	III	4	$0{,}0367 \times 2{,}37^{\pm1}$	$1{,}83 \times 4{,}24^{\pm1}$	$30{,}3 \times 1{,}93^{\pm1}$
	IV	5	$0{,}0065 \times 5{,}76^{\pm1}$	$4{,}98 \times 1{,}21^{\pm1}$	$2{,}0 \times 5{,}80^{\pm1}$

In 1950, atmospheric [129]I/[127]I ratios in background regions of the northern hemisphere exceeded 10^{-9} [1]. [129]I deposition does not show a prominent bomb peak in an alpine ice-core [26] as for instance observed for [14]C [27], [36]Cl [28] and [137]Cs [26]. Isotopic ratios and deposition densities continued to increase in Europe until the end of the 1980s (Fig. 1). Since then, ratios of nearly 10^{-6} were observed in Germany and Switzerland and remained constant until today. The fall-out of the Chernobyl accident by wet deposition was just a short-term episode with a highest measured [129]I/[127]I ratio of nearly 10^{-5}.

It appears that the [129]I/[127]I isotopic ratios are about 10^{-10} in the oceanic mixing layer remote from emittents [13,29,30]. In the English Channel, the Irish Sea, the North Sea an the North Atlantic, Raisbeck and Yiou [6,12] observed [129]I/[127]I isotopic ratios between 10^{-10} and 9×10^{-7} in samples taken between 1984 and 1992. By such isotopic ratios the results obtained by us for precipitation cannot be explained. [129]I/[127]I isotopic ratios exceeding 10^{-6} were observed by Raisbeck and Yiou [6,12] only for two samples taken close to the outlet of the La Hague plant. Since isotopic ratios between 1.4×10^{-6} and 2.4×10^{-6} were observed by us at the German coast in 1999 and 2000 it appears that the data by Raisbeck and Yiou [6,12] are no longer representative for the present status and that new systematic analyses of [129]I in the North Sea are urgently needed.

The annual [129]I deposition rates as calculated from the measured [129]I concentrations in precipitation and annual precipitation rates rose during the last five decades from 0.01 mBq m⁻² a⁻¹ in 1950 in Switzerland by nearly three orders of magnitude to a geometric mean of $6.4 \times 2.1^{\pm1}$ mBq m⁻² a⁻¹ for all four regions of Lower Saxony, Germany, during 1997 - 1999. The present deposition in Lower Saxony is inhomogeneous with time and place [17]. The fact, that [129]I emissions of the La Hague reprocessing plant, which is the dominating European source, also increased by about three orders of magnitude does not yet give conclusive evidence about the pathways of [129]I from the emittents to Germany and Switzerland. Though the majority of the emissions occurs into the sea, it is not yet clear whether the present fall-out is dominated by the marine or the atmospheric emissions [31].
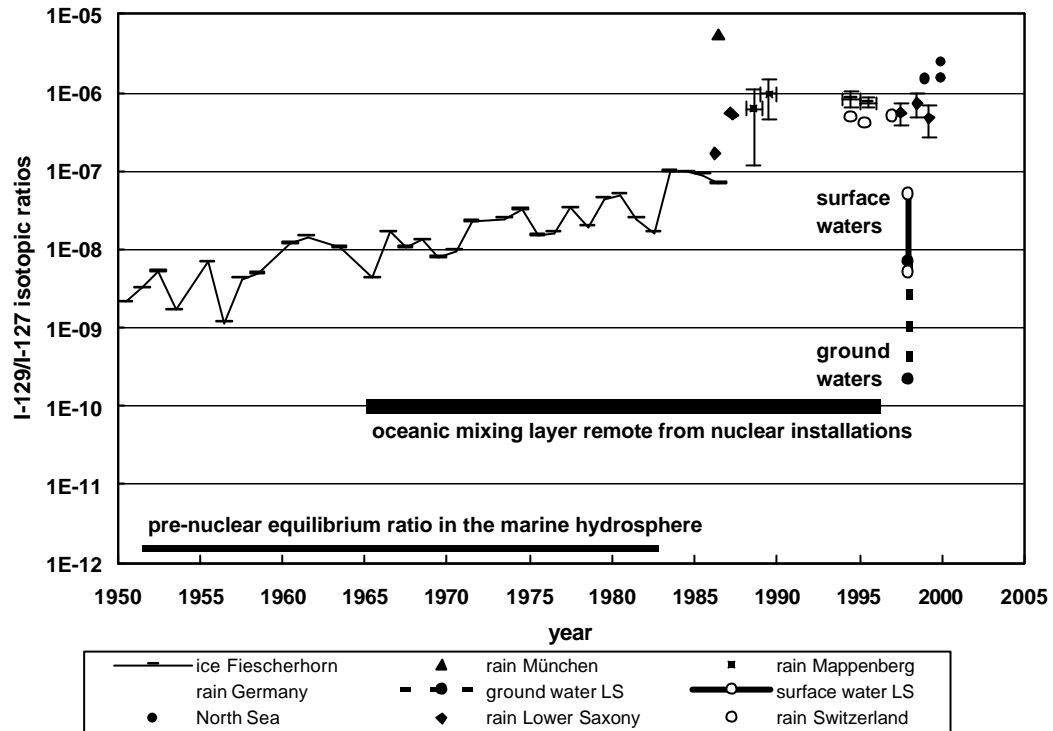
Fig. 1: Development of $^{129}I/^{127}I$ isotopic ratios in Europe as revealed by analyses of precipitation from Switzerland and Germany [18,25,26,31-35]. Data of refs. [26], [31], and [32] are calculated under the assumption of an ^{127}I content of 1,4 µg/kg.

Table 2: Annual deposition densities of ^{129}I and ^{127}I in Germany and Switzerland

location	material	year	^{127}I in mg m^{-2} a^{-1}	^{129}I in mBq m^{-2} a^{-1}	ref.
Fiescherhorn, CH	ice core	1950	-	0,014	26
		84/85	-	0,7 - 0,9	26
Mappenberg, D	rain + aerosol	88/89	-	4,0 - 6,6	32
D, 5 locations	precipitation	94/95	-	5,1 - 11	35
Dübendorf, CH	precipitation	94-97	-	2,3 - 5,3	31,34
LS, region I	rain, open field	97-99	1,6 - 2,6	8,4 - 15,6	15,16
LS, region II	rain, open field	97-99	1 - 1,6	3,4 - 6,6	15,16
LS, region III	rain, open field	97-99	0,8 - 1,9	1,9 - 6,2	15,16
LS, region IV	rain, open field	97-99	0,8 - 1,2	1,8 - 3,9	15,16
LS, region II	rain, through-fall	97-99	2,4 - 4,7	7,9 - 18,1	15,16
LS, region III	rain, through-fall	97-99	3,2 - 5,7	9,1 - 18,5	15,16

4. ^{127}I and ^{129}I in European soils

The investigation of iodine isotopes in soils is of interest for several reasons. First, the investigation of soil profiles gives clues about the migration behavior of ^{127}I and ^{129}I. Second, such data are necessary to derive and interpret integral deposition densities which are needed for balancing the anthropogenic changes or for the retrospective determination of ^{131}I fall-out after e.g. the Chernobyl accident. Third, the depth dependence of ^{127}I in soils is of interest with respect to the iodine balance in dependence on vegetation cover, soil type and characteristics. Finally, the investigation of pre-nuclear soils can give information on the natural ^{129}I/^{127}I ratios in the terrestrial biosphere.

Our investigations cover up to now pre-nuclear soil profiles from Russia [17], 44 soil profiles from Moscow and from Northern Ukraine [20,25], and 7 deep soil profiles from Lower Saxony/Germany [21]. The latter gave important clues about the iodine balance and the iodine migration in soils. Illustratively, we show in Fig. 2 the depth dependence of the surface projected concentrations of ^{129}I and ^{127}I, which usually are interpreted as deposition densities, and of the ^{129}I/^{127}I ratios in a soil profile from Klein Lobke, Lower Saxony, Germany.

Assuming that the stable iodine in soils originates mainly from atmospheric input from the oceans, the integral ^{127}I deposition density of $3,6 \times 1,6^{\pm 1}$ g m^{-2} observed in the seven soil profiles from Lower Saxony is a result of an input over thousands of years at annual ^{127}I deposition densities (table 2) of the order of 1 mg m^2 a^{-1}. The surface projected activity concentrations can, however, only be regarded as lower limits of the integral deposition densities since there is presently no quantitative information available about the loss of iodine to deeper depths or to the atmosphere by evaporation and transpiration or about the run-off by surface water. The iodine concentrations in surface and groundwater (table 1) emphasize the importance of these processes.
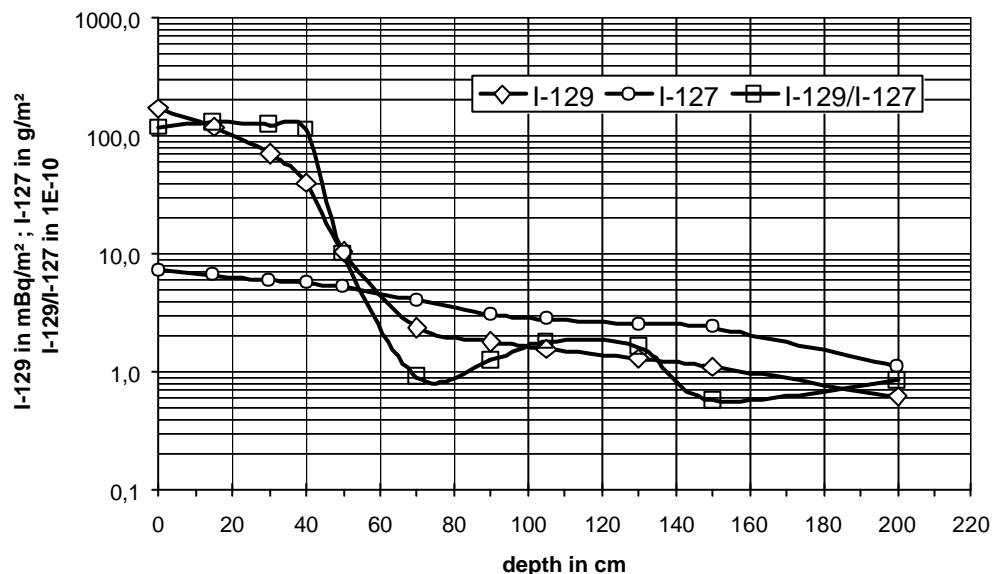
Fig. 2: Depth dependence of surface projected ^{127}I and ^{129}I concentrations and of ^{129}I/^{127}I isotopic ratios in a soil profile from Klein Lobke, Lower Saxony, Germany [21].

The problem of potential losses exists likewise for ^{129}I. However, the time scale of the man-made input is just of the order of 50 years. Taking into account the relatively low concentrations of ^{129}I and the low ^{129}I/^{127}I isotopic ratios in surface waters not affected by sea spray, i.e. in regions II-IV (table 1), one can assume these losses to be of minor importance. More than 99 % of the ^{129}I observed in the soils are located in the upper 60 cm of the profiles. But, the ^{129}I/^{127}I ratios at deeper depths still are of the order of 10^{-10}, i.e. about two orders of magnitude higher than the natural equilibrium ratio in the marine hydrosphere. This demonstrates that some man-made ^{129}I has already migrated to deeper depths.

The migration of [129]I and [127]I in the soils can only be understood as a time-dependent kinetic process involving unspecific and specific adsorption processes and isotopic exchange depending strongly on the hydrological situation [21]. As a consequence of the long time which was available for the [127]I to migrate into the ground, [127]I could occupy more stable adsorption places with long characteristic exchange times. The sorption of [129]I in the soil layers mainly occurs by non-specific sorption and the need to exchange with [127]I, which is adsorbed more strongly in deeper layers over long time scales, makes the [129]I appear to move faster than the [127]I. It turns out that the migration of the iodine isotopes cannot be described by stationary equilibrium models but need time-dependent kinetic models which are not available so far.

In spite of the uncertainty about losses of [129]I from the soil columns we calculated surface projected concentrations as a first order approximation of integral deposition densities of [129]I (table 3). The up to now lowest deposition density was found in a soil profile taken in 1939 in Lutovinovo, Russia, which also showed the lowest isotopic ratio in soils of $(0.057 \pm 0.011) \times 10^{-10}$ [16,17]. However, this ratio is a factor of four higher than the pre-nuclear marine [129]I/[127]I equilibrium ratio of $\sim 1.5 \times 10^{-12}$ [13,14]. Due to the danger of modern contamination during long-term storage, pre-nuclear soil profiles can give only an estimate of the natural equilibrium deposition densities. Contamination during sample preparation and analysis is not a problem here since the complete analytical blanks in our work typically yield [129]I/[127]I ratios below 2×10^{-13}.

Integral deposition densities in Lower Saxony, Germany, are more than three orders of magnitude higher than the pre-nuclear value derived for Lutovinovo, Russia. They are also higher than in Moscow, Russia, or Zhytomir, Ukraine, both regions being not significantly contaminated by fall-out from the Chernobyl accident. Only in areas of Ukraine which are seriously contaminated by this accident, we observed higher integral deposition densities than in Germany [21]. Generally, the [129]I deposition densities in the soil profiles from Ukraine and from Moscow show a roughly exponential decrease with depth [25]. Table 3 gives geometric means and standard deviations of integral deposition densities in contamination zones II (^{137}Cs deposition density 555 - 1480 kBq/m^2) and III (^{137}Cs deposition density 185 – 555 kBq/m^2) of Northern Ukraine. There, [129]I can be used for estimates of the fall-out of ^{131}I from the Chernobyl accident with typical geometric standard deviations of 1.5. In the highly contaminated areas of Ukraine, the majority of [129]I deposition occurred instantaneously. In Ukraine, more than 99 % of the Chernobyl fall-out still resides in the top 30 cm of the soils.

Table 3: Integral deposition densities in different regions of Germany, Russia, and Ukraine.

location	number of profiles	depth in cm	year of sampling	integral [129]I deposition densities in mBq m^{-2}
Lutovinovo, Russia	1	35	1939	0,084 ± 0,017
Moscow, Russia	2	40	1996	$49 \times 1,5^{\pm 1}$
Zhytomir, Ukraine	12	40	1997	$38 \times 1,7^{\pm 1}$
Lower Saxony/D	7	250	1999	$168 \times 1,5^{\pm 1}$
Zone III, Ukraine	24	40	1995	$130 \times 1,5^{\pm 1}$
Zone II, Ukraine	7	40	1995	$848 \times 1,5^{\pm 1}$

5. [129]I IN THE BIOSPHERE

Also the present biospheric iodine isotopic composition has been drastically changed compared to pre-nuclear values. The up-to-now lowest isotopic ratio has been measured in a pig thyroid gland powder prepared by Parke-Davis in 1943 in the USA. The mean of the two analyses of (0.058 $\pm$

0.012) $\times$ 10^{-10} [16,17] is still higher than the measured pre-nuclear ratios in ocean sediments. The thyroid glands sampled in 1947 in the USA turned out to have $^{129}I/^{127}I$ ratios 10 to 60 times higher than the pre-nuclear marine equilibrium ratio and thus are suspected to be affected already by ^{129}I releases during the Manhattan project and the onset of the nuclear age.

Present days' isotopic ratios in human and animal thyroid glands from Lower Saxony in Germany, which is far away from nuclear installations emitting ^{129}I, show much higher ratios (Fig. 3). In bovine thyroid glands, $^{129}I/^{127}I$ ratios of (110 $\pm$ 10) $\times$ 10^{-10}, (47 $\pm$ 5) $\times$ 10^{-10}, and (400 $\pm$ 196) $\times$ 10^{-10} were observed in 1978 (n = 25), 1981 (n= 22), and 1992/93 (n = 9), respectively [22,36,37]. The fall-out caused by the Chernobyl accident in Western Europe just appeared as a short-term peak in the isotopic ratios as revealed by analyses of animal thyroids from Austria and Germany [38]. Pre- and post-Chernobyl human thyroid glands from Germany analyzed at our institute showed $^{129}I/^{127}I$ ratios of (216 $\pm$ 114) $\times$ 10^{-10} (n = 13) and (320 $\pm$ 156) $\times$ 10^{-10} (n = 26), respectively. $^{129}I/^{127}I$ ratios are about one order of magnitude lower in thyroid glands from the southern hemisphere as revealed by analyses of human and animal thyroid glands from Chile [22]. This is well in line with the general differences between global weapons fall-out in the southern and northern hemispheres, on the one hand, and the fact that the emissions from reprocessing plants mainly take place on the northern hemisphere, on the other.

With respect to the radiological significance of these $^{129}I/^{127}I$ ratios, one can estimate the associated radiation exposure on the basis of a specific activity model [1]. Assuming the data of ICRP reference man for human iodine content and uptake [39] and a dose factor for a child in its first year of 2.2 $\times$ 10^{-7} Sv/Bq [40], an equilibrium $^{129}I/^{127}I$ ratio of 100 $\times$ 10^{-10} corresponds to an annual effective equivalent dose due to intake of ^{129}I of $H_{eff} \approx$ 5 nSv/a. Thus, it can be concluded that the present situation is radiological not significant, except for the closest proximity of reprocessing plants, e.g. [41]. It is to note, however, that this is just a coarse estimate which takes not into account the existing disequilibrium of ^{129}I and ^{127}I in the environment.

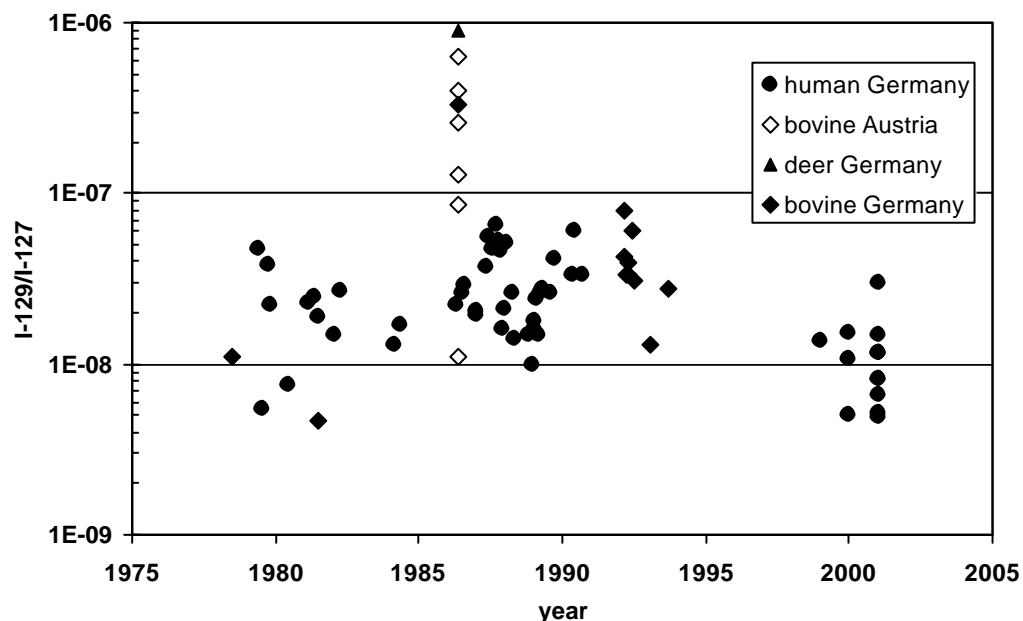Fig. 3: $^{129}I/^{127}I$ isotopic ratios in human and animal thyroid glands.

The $^{129}I/^{127}I$ ratios observed up to now in human and animal thyroid glands in Europe do not reach the ratios in precipitation (Fig. 3). Given the continuous input of ^{129}I, a further increase of biospheric $^{129}I/^{127}I$ ratios is to be expected. In the data obtained over 20 years there appeared an average increase by about an order of magnitude, though there is a considerable scatter of the data. The data of the recent thyroid glands from Lower Saxony sampled between 1999 and 2001 show slightly lower $^{129}I/^{127}I$ ratios of (132 $\pm$ 82) $\times$ 10^{-10} (n = 17) than observed previously.

Generally, the iodine isotopic ratios in the thyroids do not reflect the high ratios in the fall-out but rather the buffered system of top-soils and surface water. Consequently, the pathways of long-lived iodine is more complex than those of short-lived iodine isotopes. Since there is a strong disequilibrium of iodine isotopes in the different environmental compartments, the exposure of man and animal to [129]I, in principle, cannot be estimated on the basis of a specific activity model and the above estimate is just a zero order approximation. For a realistic assessment of the exposure to [129]I the radioecological pathways of [129]I to man and animal have to be investigated in more detail. Respective investigations of these pathways are underway.

6. CONCLUSIONS

In Europe, as all over the world, the natural abundance of [129]I have been changed in a sustainable way. But, in Western Europe these changes are going on. [129]I/[127]I ratios in precipitation stabilized after a decades-long increase at a level of nearly 10^{-6}. These ratios are more than one order of magnitude higher than recent ratios of biosphere iodine measured in Germany. The different environmental compartments in Europe show extreme disequilibrium between [129]I and stable iodine with the expected consequence of a further rise of biospheric [129]I/[127]I ratios. In spite of the fact that the present global biospheric [129]I abundance does not give rise to a significant radiation exposure, the future development should be carefully surveyed. [129]I turns out to be an outstanding quantitative indicator of the long-term human impact onto the environment. It has the potential of a powerful tracer of environmental processes and offers a possibility of retrospective dosimetry after accidental and routine fall-out situations.

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