

# <sup>129</sup>I in the environment

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## Abstract

*<sup>129</sup>I is a long-lived radionuclide ( $T_{1/2} = 15.7 \times 10^6$  years) whose presence in the environment has increased strongly since the beginning of the nuclear era. The main responsible for this are the nuclear fuel reprocessing plants of Sellafield and La Hague. Due to its tendency to enter the hydrological cycle, it has been strongly widespread especially over the Northern hemisphere. Besides, because of its biophilic behaviour, iodine can enter the food chain via the air as a result of aerial emissions. For these reasons, its importance from the point of view of Radioecology is extremely high. In this article we will present a short overview of some the traditional applications of <sup>129</sup>I in the environment.*

## Introduction

<sup>129</sup>I is a long-lived cosmogenic radionuclide ( $T_{1/2} = 15.7 \times 10^6$  years) for which the natural abundances have been altered in a significant way by human actions. Natural production processes lead to typical <sup>129</sup>I/<sup>127</sup>I isotopic ratios between  $10^{-12}$  (Kilius et al., 1992) and  $6 \times 10^{-13}$  (Roberts et al., 1997). The natural inventory of <sup>129</sup>I has been estimated to be ~230 kg, most of which resides in the deep oceans (Rao and Fehn, 1999). <sup>129</sup>I has also been produced anthropogenically during the last 60 years during fission processes associated with civil and military nuclear activities and accidents, such as Chernobyl (1986). An estimated 50–150 kg of <sup>129</sup>I were released to the atmosphere during the main period (1945–1964) of nuclear weapons testing (Wagner et al., 1996) and a further ~6 kg were released due to the Chernobyl accident (1986) (Gallagher et al., 2005).

However, the most significant contribution to the mobile <sup>129</sup>I inventory in the environment comes from succeeding activities of nuclear fuel reprocessing plants and, in particular, the liquid and gaseous releases from the two major European reprocessing facilities, namely NDA's plant at Sellafield (UK) and AREVA's plant at Cap de La Hague (France), which now account for >90% of the total global releases (Aldahan et al, 2007). Airborne releases from these facilities, and the fraction of their liquid <sup>129</sup>I releases volatilised from the ocean surface have been shown to be the main source of <sup>129</sup>I deposited over Europe.

The total annual <sup>129</sup>I discharge from both facilities remained below 20 kg year<sup>-1</sup> (0.1 TBq year<sup>-1</sup>) until the beginning of the 1990s. Later the discharge increased up to 300 kg year<sup>-1</sup> (2 TBq year<sup>-1</sup>) with 75% originating from La Hague facility (UNSCEAR, 2000). The <sup>129</sup>I discharges from these facilities account for >95% of the total inventory in the global ocean until 2000. This anthropogenic source thus predominated over all other sources including natural production, nuclear testing and other nuclear activities and accidents.

## Detection and measurement

<sup>129</sup>I decays by emitting  $\beta$ -particle with a maximum energy of 154.4 keV and  $\gamma$ -rays of 39.6 keV as well as X-rays (29–30 keV). It is possible to detect it by  $\gamma$ -spectrometry and  $\beta$ -counting by Liquid Scintillation Counting (Verrezen et al., 1992). However, these techniques show very low efficiency

because of its long half-life. A more sensitive method for the determination of  $^{129}\text{I}$  is neutron activation analysis. In this method, the sample is irradiated with neutrons in a reactor to convert  $^{129}\text{I}$  to short-lived  $^{130}\text{I}$  ( $T_{1/2} = 12.36$  h) by reaction  $^{129}\text{I}(n, \gamma)^{130}\text{I}$ , which is then measured by  $\gamma$ -spectrometry. With this technique, better detection limits ( $1 \mu\text{Bq}$ ) were reported (Hou et al., 1999).

Mass spectrometry techniques have shown to be the most powerful tools for  $^{129}\text{I}$  detection, especially in environmental samples. ICP-MS (Inductively Coupled Plasma Mass Spectrometry) has been used for  $^{129}\text{I}$  detection. However, it shows some problems associated with low sensitivity, isobaric and molecular ions interferences ( $^{129}\text{Xe}$ ,  $^{127}\text{IH}_2$ ,  $^{89}\text{Y}^{40}\text{Ar}$ ,  $^{115}\text{In}^{14}\text{N}$ ,  $^{113}\text{Cd}^{16}\text{O}$ ), memory effects, low abundance sensitivity of ICP-MS, especially isobar  $^{129}\text{Xe}$  and tailing of  $^{127}\text{I}$  (Hou et al., 2008).

Nowadays, the most widely used technique for  $^{129}\text{I}$  detection is Accelerator Mass Spectrometry (AMS). In AMS, isotopic ratios  $^{129}\text{I}/\text{I}$  are measured by mass spectrometry combined with the use of a tandem accelerator. This reduces strongly the possible background because molecules with the same mass as the isotope of interest are broken when they pass through the accelerator stripper, typically consisting on an Ar or He gas tube. Besides, the use of high energies also allows employing nuclear detectors, to discriminate between the isotope of interest and possible molecular fragments that arrive to the detector with the same  $m/q$  ratio. Even the presence of  $^{129}\text{Xe}$  is not a problem for AMS, as ions are extracted from the ion source in charge state -1, in which Xe is not stable. As a result, the detection limit is very low, but it depends on the AMS facility, as different configuration can show different background levels. In any case, typical blank isotopic ratios of  $^{129}\text{I}/\text{I}$  in the order of  $10^{-14}$  and  $10^{-13}$  are reported (Hou & Roos, 2008). Then, AMS is the only method for the determination of  $^{129}\text{I}$  in the pre-nuclear age samples ( $^{129}\text{I}/^{127}\text{I} < 10^{-10}$ ).

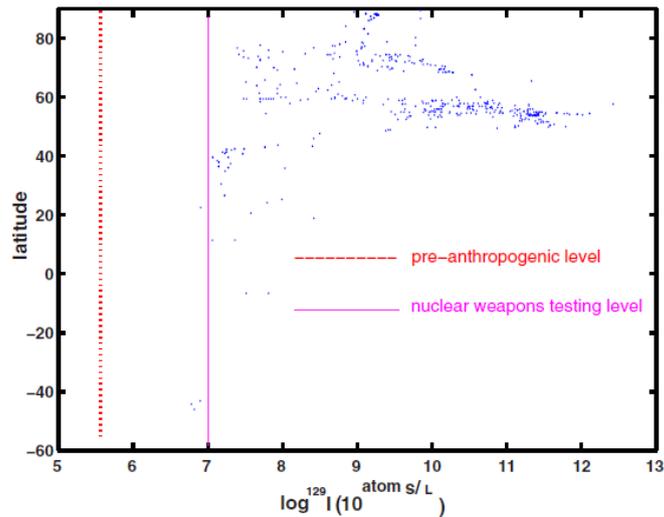
## Applications

### *$^{129}\text{I}$ in the marine environment*

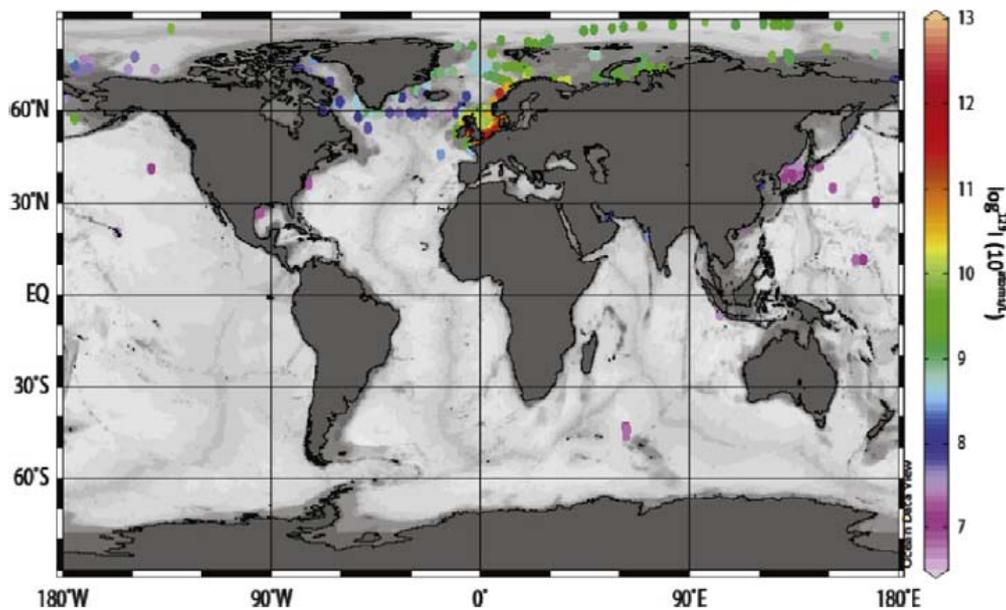
Most of the mobile iodine resides in oceans. For that reason, special attention has been paid to  $^{129}\text{I}$  as a tracer of hydrological processes in the marine environment. However, in spite of the interest and the numerous publications on the presence of this radionuclide in the seas, the picture is not complete, and more data are needed to reconstruct the information on the sources and the  $^{129}\text{I}$  activities, especially in the Southern Hemisphere.

A very good view of the scene was given in He et al. (2013). There, the authors compile a set of data published up to the moment on the presence and evolution of  $^{129}\text{I}$  in the oceans. Figure 1 shows a summary of the  $^{129}\text{I}$  concentrations in seawater samples taken all over the world as a function of latitude. It can be seen that the pre-anthropogenic level has been strongly surpassed due to the artificial discharges. While nuclear fallout due to nuclear weapons testing would be responsible of levels in the order of  $10^7$  atoms/l, concentrations of even  $10^{12}$  atoms/l have been measured in the Northern Hemisphere, for example in the Irish Sea. The main responsible for this increase are the nuclear fuel reprocessing plants of Sellafield and La Hague, as described in the introduction. Fortunately, good information on the history of the discharges is available.

Most of this  $^{129}\text{I}$  follows a well-known path. The currents take it from La Hague to the North Sea along the English Channel where it meets the  $^{129}\text{I}$  from Sellafield. This has a longer way as it surrounds the whole Scotland and the English coasts to the North Sea. Then, the currents carry this  $^{129}\text{I}$  to the Arctic Ocean, following the Norwegian Coastal Current. For that reason, these zones have been extensively studied, as shown in figure 2.



**Figure 1.** Global  $^{129}\text{I}$  marine distribution as a function of latitude (He et al., 2013).



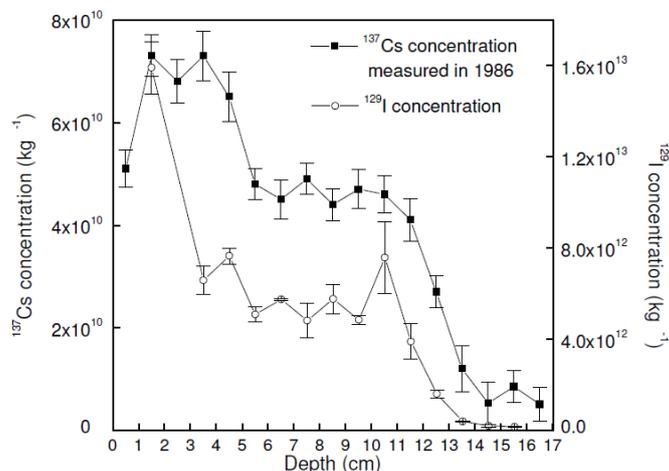
**Figure 2.** Sampling locations of published available  $^{129}\text{I}$  data and the concentrations of  $^{129}\text{I}$  in the surface water in the sampled sites (compilation by He et al., 2013).

As it can be seen, the amount of information on the presence of  $^{129}\text{I}$  in the Southern hemisphere is, however, very scarce, and even in mid latitudes in the Northern hemisphere. For this reason, more information is needed to reconstruct the behaviour of  $^{129}\text{I}$  in the global oceans.

### *$^{129}\text{I}$ in sediments*

As  $^{129}\text{I}$  has been released to the marine environment in such large quantities, it is clear that part of it has precipitated and can be found in the sediments, even when it is very conservative in water. This way, its presence in sediments can give us information not only on the amount of the contamination but also on the history of the sedimentation process and possibly on the source of the releases. An example was published by López-Gutiérrez et al. (2004), as shown in figure 3. In that work, a sediment core from the Kattegat area in the North Sea was analysed. The  $^{129}\text{I}$  results were compared

to the  $^{137}\text{Cs}$  activities measured in 1986. In both cases a strong increase at about 13 cm depth is consistent with the beginning of the operations at the nuclear fuel reprocessing plants of Sellafield and La Hague, in the 20<sup>th</sup> century. Thanks to the comparison with the history of the nuclear discharges from these plants, a lot of data on the transfer times and factors, sedimentation rates, residence time in water, and behaviour in the sediment column could be extracted.



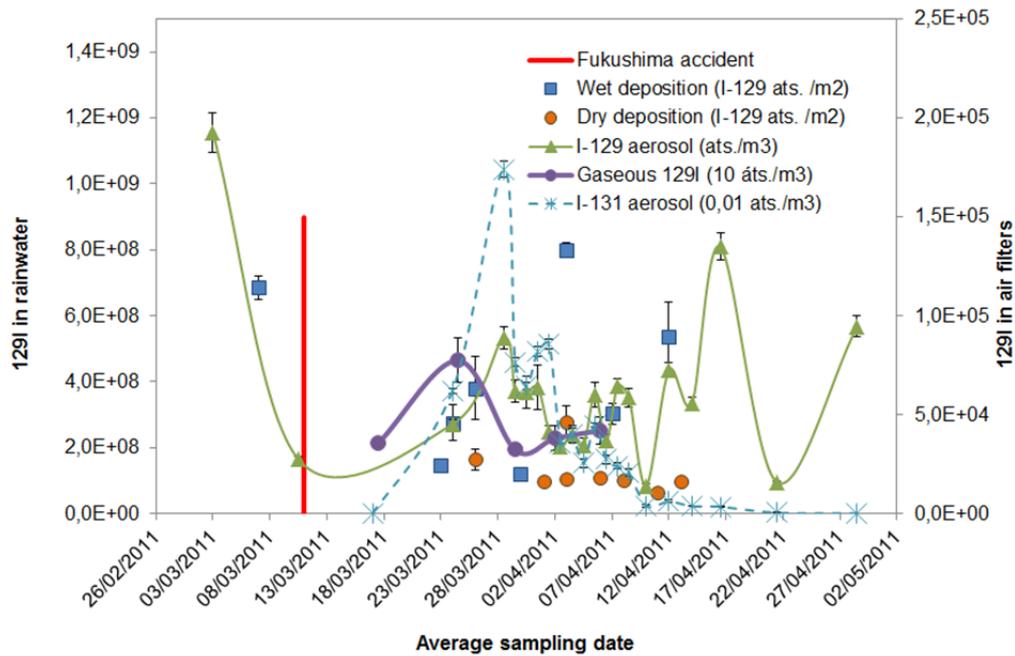
**Figure 3.** Depth profiles of  $^{137}\text{Cs}$  and  $^{129}\text{I}$  in a sediment core from Kattegat (López-Gutiérrez et al., 2004).

### *$^{129}\text{I}$ in the atmosphere*

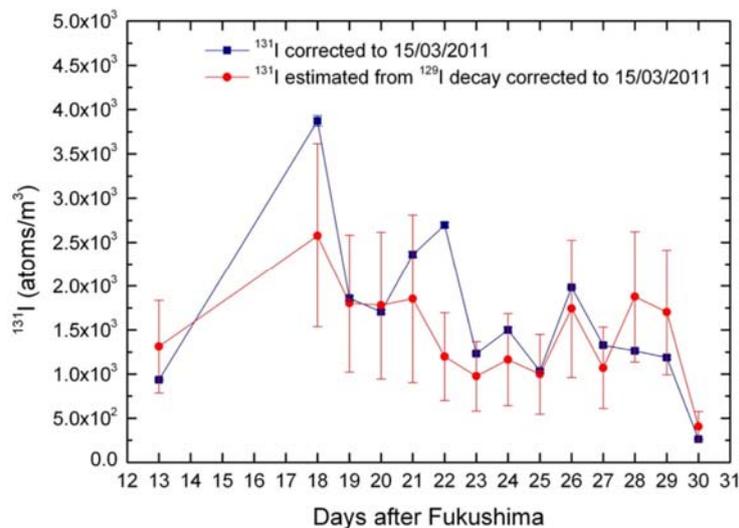
Iodine is a very volatile element. Most part of the iodine in the atmosphere is evaporated from the oceans and it remains typically around 20 days both in gaseous or particulate forms. The oceans are also a very important source of  $^{129}\text{I}$  to the atmosphere. For example, Krupp and Aumann (1999) measured isotopic ratios in the order of  $^{129}\text{I}/^{127}\text{I} \sim 10^{-7}$  in Germany in 1994 and 1995, several orders of magnitude over the pre-nuclear level of  $10^{-12}$ .  $^{129}\text{I}$  discharge by the reprocessing plants in the North Sea was probably the main responsible of these high levels.

But  $^{129}\text{I}$  is also released in a gaseous form by the nuclear fuel reprocessing plants, so the signal is very much affected by the evolution of the gaseous releases, which travel very fast long distances in a few days. Something similar happens in the case of accidental releases like the one in Fukushima in 2011. Figure 4 shows the evolution of different magnitudes in Sevilla (Spain) during the days after the accident (Gómez-Guzmán et al., 2017). It can be seen that both  $^{131}\text{I}$  and  $^{129}\text{I}$  were detected in air filters as well as in rainwater. However, the levels of  $^{129}\text{I}$  before and after the accident were even higher than during the days that  $^{129}\text{I}$  and  $^{131}\text{I}$  from Fukushima arrived to Spain. As the authors showed, the prevalent winds during those days were favourable to the arrival of Fukushima contaminated air instead of the releases from Sellafield and La Hague.

A very relevant of  $^{129}\text{I}$  in these cases, thanks to its long half-life, is that the analysis can be carried out many years after the samples were taken. In the previous example,  $^{131}\text{I}$  was measured right after the sample collection but  $^{129}\text{I}$  was measured by AMS in 2015. And then,  $^{129}\text{I}$  can help reconstruct the  $^{131}\text{I}$  impact (which, from the radiation protection is more relevant) long time after the emissions. An example of this can be seen on figure 5.



**Figure 4.**  $^{131}\text{I}$  and  $^{129}\text{I}$  detected in atmospheric samples in Sevilla (Spain) after the Fukushima accident (Gómez-Guzmán et al., 2017).



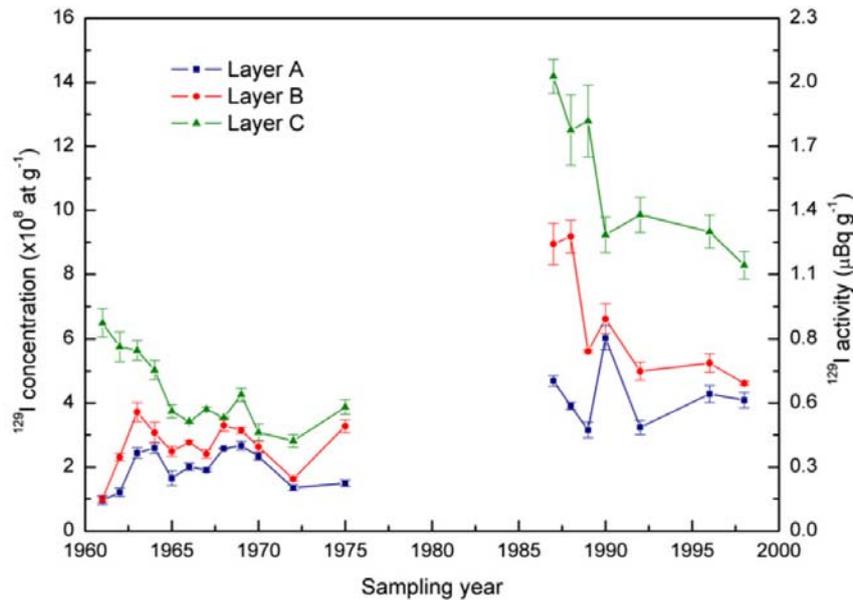
**Figure 5.** Comparison between the measured  $^{131}\text{I}$  concentration in air in Sevilla after the Fukushima accident and the estimation made through the  $^{129}\text{I}$  measurement (Gómez-Guzmán et al., 2017).

### $^{129}\text{I}$ in biota

As iodine is also a very biophilic element,  $^{129}\text{I}$  is clearly also incorporated to the living organisms. This is relevant from the Radioecology point of view itself and also from the information that can be extracted on the sources and the ways it is transported.

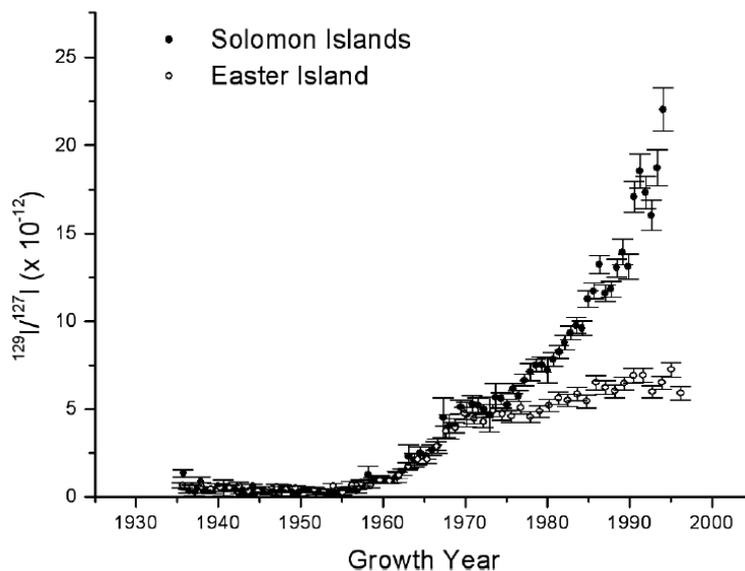
Many examples can be found in the literature. In 2011, Gómez-Guzmán et al. (2011) measured the concentration of  $^{129}\text{I}$  in lichens (*Cladonia alpestris*) from Sweden. These lichens are commonly part of the diet of reindeer, whose meat is afterwards eaten by humans. In figure 6, it is shown how  $^{129}\text{I}$  is not uniformly distributed in the different layer of the lichens. Also, the concentrations measured

after the Chernobyl accident in 1986 increased strongly with respect to the ones detected before 1975.



**Figure 6.**  $^{129}\text{I}$  in the different layers of lichen *Cladonia alpestris* from Sweden (Gómez-Guzmán et al., 2011).

Another example can be found in Biddulph et al. (2006). The authors measured  $^{129}\text{I}$  in coral skeletons from the Solomon Islands and Easter Island (figure 8), showing the increase of the levels due to the history of atmospheric nuclear weapons tests in this ocean in the 20<sup>th</sup> century and demonstrating that Coral skeletons preserve an iodine record in the tropical surface waters of the ocean.



**Figure 8.** Isotopic ratio  $^{129}\text{I}/^{127}\text{I}$  ratio in corals from the South Pacific (Biddulph et al., 2006).

As a summary, it has been shown how  $^{129}\text{I}$  is a very interesting radionuclide from the environmental point of view. Its strong value as radiological and environmental tracer is founded on a well-known history of the sources and its ability to disperse over many environmental compartments.

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