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Iodine-129 and iodine-127 in European seawaters and in precipitation from Northern Germany

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ABSTRACT

In order to obtain a comprehensive survey on the consequences of the marine ^{129}I discharges from the European reprocessing plants La Hague and Sellafield, the distribution of ^{129}I and ^{127}I in surface waters of the North Sea, the English Channel, the Irish Sea, and the Northeast Atlantic was studied using accelerator mass spectrometry for ^{129}I and ICP-MS for ^{127}I. Samples of seawater were taken in the German Bight in May, September, and November 2005 and in the entire North Sea and the English Channel in August 2005. Further samples were obtained from the Irish Sea in June and August 2006 and from Arctic waters between Spitsbergen and Southern Norway in September 2005.

^{129}I is a conservative tracer in seawater. The concentrations of ^{127}I are relatively constant with exceptions of coastal areas with high biological activity and of areas influenced by influx from rivers and the Baltic Sea. The variability of the ^{129}I/^{127}I isotopic ratios is exclusively determined by admixture of ^{129}I released from the reprocessing facilities Sellafield and La Hague to the seawater. The ^{129}I/^{127}I ratios were between $4\times10^{-9}$and 3×10^{-6}: at least 3 orders of magnitude higher than the natural equilibrium isotopic ratio 1.5×10^{-12}. ^{129}I/^{127}I ratios of a few times 10^{-10} were only found in seawater from the Indian Ocean and from the Pacific at Hawaii. Comparison of the results obtained for seawater with those of a measurement of airborne iodine species and with iodine isotopes in precipitation in Northern Germany demonstrates the transfer of ^{129}I and ^{127}I from the sea into the atmosphere and the dominating role of the marine discharges for the atmospheric fallout of ^{129}I in Western Europe. The results are discussed with the goal to estimate the relevance of the marine discharges for the contamination of the continental areas.

1. Introduction

1.1. ^{129}I in the Environment

The natural abundances of the long-lived ^{129}I ($T_{1/2}=15.7$ Ma) have been changed in a sustainable way by the fallout from nuclear weapon explosions, nuclear accidents and discharges from reprocessing plants. In nature, ^{129}I is produced by spontaneous fission of uranium and by interaction of galactic cosmic ray particles with xenon in the Earth's atmosphere. As described e.g. by Schmidt et al. (1998) in detail, the total ^{129}I-inventory of the Earth is estimated to be 50,000 kg (326.8 TBq). Most of this inventory is bound in the lithosphere and just 263 kg (1.7 TBq) make up the “free” inventory of atmosphere, hydrosphere and biosphere. The main natural sources of free ^{129}I are releases from the lithosphere through volcanic activity and production by cosmic radiation (about 45% each). In the atmosphere, hydrosphere and biosphere, ^{129}I mixes with stable ^{127}I. More than 99% of the free ^{127}I, about 8×10^{14} kg, are contained in the oceans and young oceanic sediments. Most authors agree that the ^{127}I in the atmosphere and biosphere originates mainly from these sources (Miyake and Tsunogai, 1963; Kocher, 1981; Whitehead, 1984).

The natural equilibrium isotopic ratio ^{129}I/^{127}I was estimated to be in the range of $(0.04–3.0)\times10^{-12}$ (Edwards, 1962; Edwards and Rey, 1968; Kohman and Edwards, 1966). Using a detailed equilibrium model Fabryka-Martin et al. (1984) calculated an isotopic ratio of 5.5×10^{-13} for the marine hydrosphere and for soils and the terrestrial biosphere a slightly higher ^{129}I/^{127}I-ratio of 6.5×10^{-13}. Measurements of young oceanic sediments yielded a value of $^{129}\mathrm{I}/^{127}\mathrm{I}=\sim1.5\times10^{-12}$ (Fehn et al., 1986; Schink et al., 1995; Moran et al., 1998).

The anthropogenic production of ^{129}I started with the military and peaceful use of nuclear fission. The atmospheric nuclear explosions added between 43 kg (0.28 TBq) and 150 kg (0.98 TBq) to the free ^{129}I inventory (Carter and Moghissi, 1977; UNSCEAR, 1982; Chamberlain,

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1991; Eisenbud and Gesell, 1997). The uncertainty of these estimates results from the lack of knowledge about the total fission yields of the atmospheric explosions.

The main man-made contribution to ^{129}I in the environment results, however, from the reprocessing of spent nuclear fuel. Comprehensive information about the European reprocessing plants La Hague in France and Sellafield in UK is available (Fig. 1). At Sellafield the reprocessing and consequently the discharges started already in 1951, while La Hague started in 1966. For the early years of Sellafield only data for the atmospheric discharges are available. Until the year 2005 the Sellafield and La Hague plants discharged about 4920 kg (32.2 TBq) ^{129}I (Raisbeck et al., 1995; Gray et al., 1995; Parker, 2001; Webmaster Cogema, 2001; BNFL, 1999; Groupe Radioécologie Nord-Cotentin, 1999; Cogema, 2003; Areva, 2003, 2005; Environment Agency et al., 2005, 2006). About 70% of the total ^{129}I discharges were from La Hague. The majority of these discharges, about 85% of the ^{129}I from Sellafield and about 97% of the ^{129}I from La Hague, were marine discharges into the Irish Sea between Ireland and Great Britain and the English Channel between Great Britain and the continent, respectively.

The total atmospheric discharges up to 2005 were 140 kg (0.9 TBq) for Sellafield and 69 kg (0.45 TBq) for La Hague and the total marine discharges were 1380 kg (9.0 TBq) for Sellafield and 3330 kg (21.8 TBq) for La Hague. These estimates are in reasonable agreement with those of Aldahan et al. (2007) and Reithmeier et al. (2010), though some differences for the atmospheric discharges from Sellafield exist.

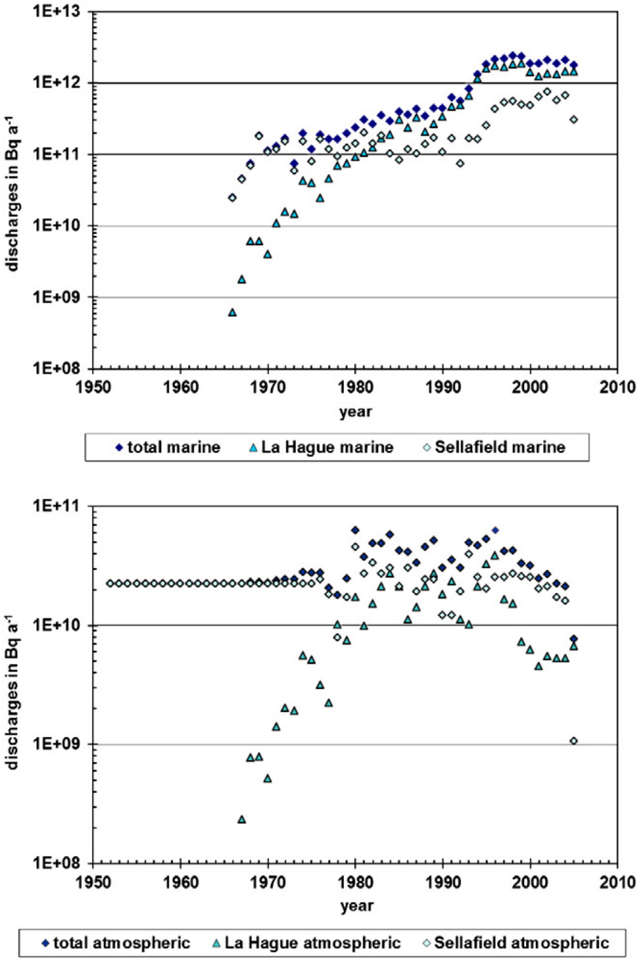

Fig. 1. Temporal development of the marine (top) and atmospheric (bottom) discharges of ^{129}I from the European reprocessing plants La Hague and Sellafield. The data until 1994 were reconstructed by Raisbeck et al. (1995); since 1994 measured data are available (see text for references).

The reprocessing plant at Marcoule in France also discharged significant amounts of ^{129}I into the European environment. Marcoule started in 1959 and ended in 1997. Atmospheric discharges of 68 kg (0.45 TBq) were estimated by Aldahan et al. (2007), while Reithmeier et al. (2010) gave 184 kg (1.2 TBq) for Marcoule. There are no quantitative estimates of the discharges from Marcoule into the Rhone River and ultimately into the Mediterranean Sea. Yiou et al. (1997) investigated ^{129}I in water from the Western Mediterranean Sea and from the Rhone Estuary. From these data they concluded that the liquid discharges from Marcoule were less than 1% of the marine discharges from Sellafield and La Hague. The impact on the Mediterranean Sea was confined to the proximity of the Rhone Estuary, where ^{129}I/^{127}I isotopic ratios of about 5×10^{-7} were observed. The other samples from the Mediterranean showed ^{129}I/^{127}I isotopic ratios less than 10^{-8}. In the context of this work, the influence of the liquid discharges from Marcoule on the contamination situation in Northern Germany can be neglected.

Up to now, there are just very limited reports about the discharges from eastern and western military reprocessing plants (Moran et al., 1999; DeWayne et al., 2003; Hu et al., 2005; Reithmeier, 2005; Snyder et al., 2010). The reactor accident of Chernobyl just added a relatively small amount to the free ^{129}I inventory. About 2 kg (13 GBq) and about 6 kg (40 GBq) were estimated by Schmidt et al. (1998) and Aldahan et al. (2007), respectively.

The discharges from the European reprocessing plants, La Hague and Sellafield, show considerable differences, both with respect to their contributions to the different types of discharges and to their temporal dependences (Fig. 1). The atmospheric discharges from Sellafield stayed constant within an order of magnitude since 1952, while those from La Hague increased from 1966 to 1996 by more than two orders of magnitude and then dropped again by nearly an order of magnitude, being, however, mostly much lower than those from Sellafield.

The total marine discharges increased from 1966 nearly continuously until 1995 and then levelled off and even slightly decreased since 2000. In the beginning, the discharges from Sellafield exceeded those from La Hague, in 1986 La Hague broke even, and since that time the La Hague marine discharges dominate. However, during recent years a renewed increase of the marine discharges is also seen in Sellafield.

Man-made ^{129}I has changed the natural isotopic abundances of iodine globally and locally. The ^{129}I/^{127}I isotopic ratios have been increased in the oceanic mixed layer by about two orders of magnitude to ^{129}I/^{127}I ~ 10^{-10} (Kilius et al., 1994). In the Irish Sea and in the Channel ^{129}I/^{127}I ratios of up to 3.7×10^{-6} were reported for samples of water and seaweed taken in 1992 (Yiou et al., 1994).

The atmospheric discharges of ^{129}I from Sellafield and La Hague are transported with the atmosphere and lead to a widespread contamination of the environment in Western Europe. The marine discharges from Sellafield spread around the Irish Sea. This ^{129}I is then further transported with the water masses northerly around Scotland, enters the North Sea, and is mixed with the water current coming out of the Channel, which carries the marine discharges from La Hague, along the Belgian, Dutch, German, and Danish shores. The ^{129}I is transported along the Norwegian coast into the Nordic Seas (Greenland, Iceland and Norwegian Seas) and the further into the Arctic Ocean. The main ^{129}I contamination in the Arctic Ocean originates from discharges of the European reprocessing plants (Smith et al., 1998, 1999; Buraglio et al., 1999; Alfimov et al., 2004a; Yiou et al., 1994; Yiou, 2002). For detailed descriptions of the water currents in the Northeast Atlantic, the English Channel and the North Sea see Dahlgaard (1995) and Keogh et al. (2007).

The contamination of the oceans also causes contamination of the continents. Iodine is released from the sea surface mainly via CH$_3$I, transported with the air over land where it precipitates as dry or