



Iodine-129, Iodine-127 and Caesium-137 in the environment: soils from Germany and Chile

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ABSTRACT

Soil profiles from Bavaria in southern Germany and from Chile were analysed for ^{129}I by accelerator mass spectrometry (AMS), for ^{127}I by inductively coupled plasma mass spectrometry (ICP-MS), and for ^{137}Cs by gamma-spectrometry. The mean deposition density of ^{137}Cs in soils from Bavaria was $(41 \times 1.5^{\pm 1}) \text{ kBq m}^{-2}$ (geometric mean and geometric standard deviation), originating mostly from the Chernobyl fall-out. The deposition density of ^{129}I in these soils was $(109 \times 1.5^{\pm 1}) \text{ mBq m}^{-2}$. The dominant sources of ^{129}I in Bavaria are, however, the reprocessing plants La Hague and Sellafield and not the Chernobyl fall-out. The $^{129}\text{I}/^{127}\text{I}$ isotopic ratios of the Bavarian soils were between 10^{-7} and 10^{-10} , i.e. 10^2 – 10^5 times higher than the ratios observed for the samples from Chile. The ^{129}I integral deposition densities in Chile, Easter Island and Antarctica were between 0.3 mBq m^{-2} and 2 mBq m^{-2} . In these soils, the observed $^{129}\text{I}/^{127}\text{I}$ ratios were about 10^{-12} . The soils from Chile allow the determination of the ^{129}I fall-out from the atmospheric nuclear weapons explosions undisturbed from contaminations due to releases from reprocessing plants. An upper limit of the integral ^{129}I deposition density of the atmospheric nuclear weapons explosions on the Southern Hemisphere (27°S) is about 1 mBq m^{-2} . Finally, the dependence of the migration behaviour of ^{137}Cs , ^{127}I and of ^{129}I on the soil properties is discussed. It turns out that there is a distinctly different behaviour of ^{127}I , ^{129}I , and ^{137}Cs in the soils exhibiting different sorption mechanisms for old and recent iodine as well as for ^{137}Cs .

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1. Introduction

The long-lived radionuclide ^{129}I ($t_{1/2} = 15.7 \text{ Ma}$) is naturally produced by cosmic-ray induced spallation of xenon in the atmosphere and by spontaneous fission of ^{238}U . The total natural ^{129}I inventory of the geosphere was estimated by Fabryka-Martin (1982) to be 50,000 kg (326.8 TBq); see Schmidt et al. (1998) for details and further references. Most of this inventory is bound in the lithosphere and just 263 kg (1.7 TBq) is the “mobile” inventory of atmosphere, hydrosphere and biosphere. The main natural sources of mobile ^{129}I are releases from lithosphere through volcanic activity and production by cosmic radiation (about 45% each). In atmosphere, hydrosphere and biosphere, ^{129}I mixes with stable ^{127}I and enters the environmental iodine cycle. Over long time scales, equilibrium between ^{129}I and ^{127}I can be assumed in the mobile inventory.

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The natural equilibrium of the $^{129}\text{I}/^{127}\text{I}$ isotopic ratios was estimated to be in the range of $(0.04\text{--}3.0) \times 10^{-12}$ (Edward, 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 $^{129}\text{I}/^{127}\text{I}$ isotopic ratio of 6.5×10^{-13} . Measurements of young oceanic sediments yielded a value of $^{129}\text{I}/^{127}\text{I} = \sim 1.5 \times 10^{-12}$ (Fehn et al., 1986; Schink et al., 1995; Moran et al., 1998). A pre-nuclear equilibrium $^{129}\text{I}/^{127}\text{I}$ isotopic ratio of $(2.0 \times 1.4^{\pm 1}) \times 10^{-13}$ in the continental biosphere was proposed on the basis of pre-nuclear iodine preparations (Ernst, 2003; Ernst et al., 2003).²

More than 99% of the mobile ^{127}I , about $8 \times 10^{14} \text{ kg}$, is contained in the oceans and young oceanic sediments (Fabryka-Martin et al., 1984). According to Muramatsu and Wedepohl (1998) most of the iodine in the Earth's crust is contained in marine sediments (68%) and only 0.8% is dissolved in the seawater. Igneous and metamorphic rocks contain only 2.7%, marine basalts 0.6% and

² We use the convenient notation of $2.0 \times 1.4^{\pm 1}$ to present geometric means, here 2.0, and geometric standard deviations, here 1.4.

sedimentary rocks 28%. Most authors agree that the ^{127}I in the atmosphere and biosphere originates mainly from marine sources (Miyake and Tsungai, 1963; Kocher, 1981; Whitehead, 1984).

Iodine is released not only from the ocean surface mainly as CH_3I , but also as I_2 and HOI . A complex photochemistry oxidises the iodine species to I^- and IO_3^- . The atmospheric iodine makes its way to the continents where it is deposited by wet and dry deposition. The precipitation interacts with soils and the iodine is partially adsorbed in the soils. Surface and groundwater transport iodine back into the oceans. A complicated compartment system with highly differing time scales must be assumed to describe the pathways in the global mobile iodine inventory. Locally, the situation is even more complicated. On the way of the air masses over the continents, the atmospheric iodine is depleted resulting in lower deposition rates deep inside the continents compared to ocean-near areas. See Fabryka-Martin et al. (1984), Yoshida and Muramatsu (1995), Cox et al. (1999), Amachi et al. (2001), Muramatsu et al. (2004), Baker (2005), Pechtl et al. (2007), O'Dowd and Leeuw (2007), and Englund et al. (2010) for further details and discussions.

Iodine in surface water and soils are the essential sources of iodine in the terrestrial biosphere. Iodine is a biophilic element and enriched in organic materials such as proteins, polyphenols and humic substances. Large amounts of ^{127}I and ^{129}I in freshwater, estuaries and oceanic surface waters are associated with macromolecular organic matter (Schall et al., 1997; Moore and Grosko, 1999; Schwehr and Santschi, 2003).

Man-made ^{129}I has been released into the environment by atmospheric atomic explosions, nuclear fuel reprocessing facilities and nuclear reactor accidents. Nuclear fuel reprocessing is the important source of the man-made ^{129}I into the environment. As discussed in detail elsewhere (Michel et al., 2012), estimates of the ^{129}I releases from atmospheric atomic explosions range from 43 kg (0.28 TBq) to 150 kg (0.98 TBq) and of the Chernobyl accident from 2 kg (13 GBq) to 6 kg (40 GBq). Up to the year 2005, the European reprocessing plants, La Hague and Sellafield discharged 251 kg (1.64 TBq) into the atmosphere and 4720 kg (30.9 TBq) into the English Channel and the Irish Sea, respectively. For the reprocessing plant Marcoule estimates of the atmospheric discharges are between 68 kg (0.45 TBq) and 184 kg (1.2 TBq). No estimates of the discharges into the Rhone River are available for Marcoule. For the other, in particular the military, reprocessing plant only few data exist, so that any estimate of the global releases remains incomplete.

Human nuclear activities and the associated releases of man-made isotopes are heavily concentrated in the Northern Hemisphere. Only three nuclear power plants and two reprocessing plants are located in the Southern Hemisphere. The reprocessing plants in the Northern Hemisphere are dominating the global ^{129}I inventory and more than 95% of the present mobile ^{129}I is distributed in the Northern Hemisphere (Snyder et al., 2010). Fehn and Snyder (2000) reported that the concentrations of ^{129}I in the Southern Hemisphere are lower by about two orders of magnitude than in the Northern Hemisphere.

Man-made ^{129}I was globally distributed in the environment and as a consequence the natural equilibrium isotopic $^{129}\text{I}/^{127}\text{I}$ ratios were increased by several orders of magnitude. The $^{129}\text{I}/^{127}\text{I}$ ratios in the atmosphere and in precipitation increased from a few times 10^{-12} to 10^{-9} between 1940 and 1950 in background regions of the Northern Hemisphere. In 1950, atmospheric $^{129}\text{I}/^{127}\text{I}$ ratios exceeded 10^{-9} and today they have, for instance, reached 10^{-6} such as in European seawater and north Atlantic. For more details see Yiou et al. (1994, 2002), Raisbeck et al. (1995), Fehn and Snyder (2000), Hou et al. (2002), Buraglio et al. (2000), López-Gutiérrez et al. (2004), Alfimov et al. (2004), Michel et al. (2005b, 2012),

Schnabel et al. (2007), Aldahan et al. (2007), Reithmeier et al. (2010) and Snyder et al. (2010).

The development with time of the ^{129}I deposition rates in Western Europe such as in Germany and Switzerland are known from the analyses of an alpine ice core and from direct measurements; see Michel et al. (2005b, 2012) for data and further references. The ^{129}I deposition rates derived from the Fiescherhorn ice core (Wagner et al., 1996) do not show a prominent bomb peak as for instance observed for ^{36}Cl and ^{137}Cs in the same ice core (Synal et al., 1990). Up to now, only one profile of Mississippi River Delta sediments showed a “bomb peak” for ^{129}I on the Northern Hemisphere, in parallel with one of plutonium isotopes (Oktay et al., 2000). However, due to the complex and unknown input function of these sediments no total deposition inventory of the “bomb peak” was estimated by these authors.

Due to the releases pattern, atmospheric isotopic ratios and ^{129}I deposition rates continued to increase in Europe until the end of the 1980s. Since then, ratios of nearly 10^{-6} were observed in Germany and Switzerland and remained quite constant until today (Ernst, 2003; Ernst et al., 2003; Michel et al., 2005b, 2012). The fall-out of the Chernobyl accident, mainly by wet deposition, was just a short-term episode with a highest measured $^{129}\text{I}/^{127}\text{I}$ ratio of nearly 10^{-5} (Paul et al., 1987). Englund et al. (2008) investigated a sediment profile from Sweden in an area highly contaminated by Chernobyl fall-out, but because of the dominating signal from the reprocessing plants the Chernobyl contribution to ^{129}I was not as clear as in the case of ^{137}Cs . Nowadays the North Sea appears to be the dominant source of air-borne iodine in Northern Europe due to the emissions of European reprocessing plants (Michel et al., 2005b, 2012; Nies et al., 2010).

In contrast to ^{129}I , the natural production of ^{137}Cs ($t_{1/2} = 30.07$ Ma) by spontaneous fission does not cause any significant natural abundances in the atmosphere, hydrosphere or biosphere because of its relatively short half-life. Man-made ^{137}Cs , however, has an important impact on the human environment. It started with the atmospheric explosions of atomic bombs which released about 948 PBq ^{137}Cs (UNSCEAR, 2000). During the Chernobyl accident 85 PBq ^{137}Cs were set free (UNSCEAR, 2011). The releases of ^{137}Cs from reprocessing plants are comparably low and those from nuclear power plants in normal operation even negligible. Releases of ^{137}Cs from Sellafield and La Hague were mainly marine ones: 1 PBq from La Hague and 41 PBq from Sellafield until 2005 (OSPAR, 2008). Atmospheric releases of ^{137}Cs from La Hague were less than 1 GBq for La Hague and less than 3.4 TBq for Sellafield up to 1997 (UNSCEAR, 2000, Annex C). Data of ^{137}Cs releases into the atmosphere for other reprocessing plants are not available. Also for nuclear power plants in normal operation no comprehensive data on releases into the atmosphere exist. Upper limits of ^{137}Cs releases can, however, be estimated from the release data for radioactivity bound to particulates demonstrating their insignificance. The geometric mean of the normalised release of particulate bound radioactivity is about 10^{-2} GBq/GWa to a few times 10^{-1} GBq/GWa (UNSCEAR, 2000, Annex C). Therefore, we can restrict our discussion on ^{137}Cs from atmospheric explosions and from the Chernobyl accident.

^{137}Cs – as well as ^{129}I – from the atomic explosions was globally distributed by stratospheric transport and deposited by long-term fall-out. Due to the global atmospheric transport processes (UNSCEAR, 1982) the bomb fall-out depended strongly on the geographical latitude. Decay corrected to 1981, the bomb fall-out of ^{137}Cs was about 5 kBq m^{-2} at northern latitudes between 40° and 60° and about a factor of 5 lower at the same southern latitudes. According to UNSCEAR (1982) about 25% of the global ^{137}Cs fall-out was deposited on the Southern Hemisphere. Schuller et al. (2002) measured deposition densities of ^{137}Cs in Chilean soils between 0.5 kBq m^{-2} and 5.4 kBq m^{-2} , the latter figure being surprisingly

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