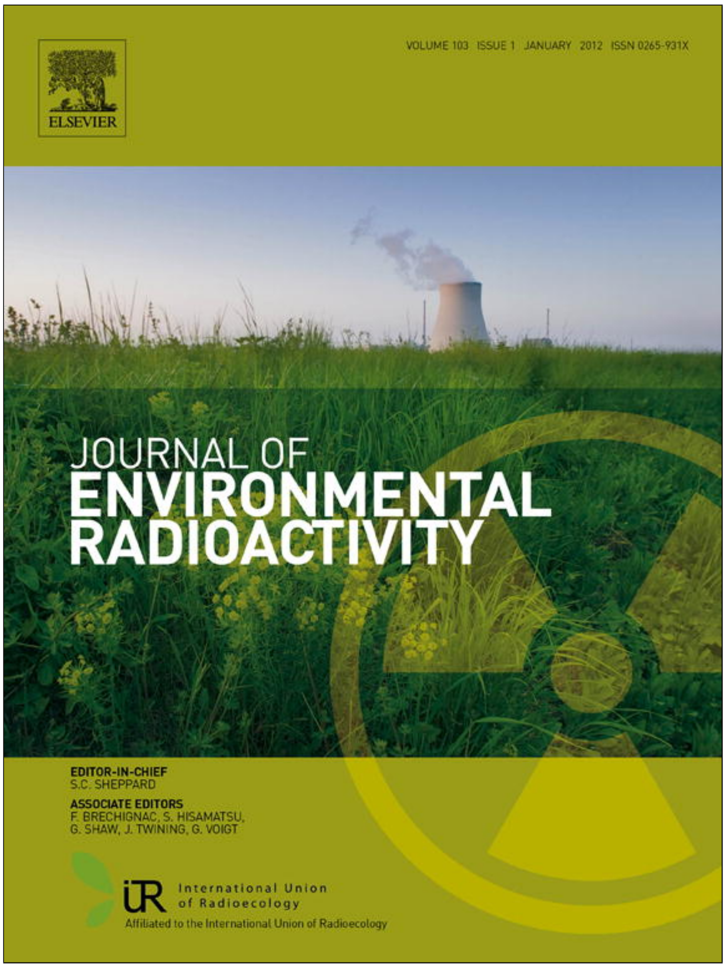
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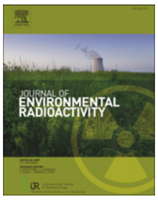

Retrospective measurements of airborne ^{129}Iodine in Austria

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ABSTRACT

The knowledge about the distribution of anthropogenic ^{129}I is crucial for a successful establishment of transport mechanisms, fate and behaviour in the environment. In present study, the historical record of dry deposition of ^{129}I in Austria over four decades back to the 1960s is reconstructed. The ^{129}I/^{127}I isotopic ratio of the order of 10^{-9}–10^{-7} in airborne particles revealed a prominent anthropogenic ^{129}I signature. The time profile of airborne ^{129}I follows directly the pattern of the gaseous emissions from European reprocessing plants. Furthermore, temporal variations of ^{129}I were traced monthly over two years. The potential risks of internal exposure to ^{129}I are associated with both inhalation and ingestion. Since dose via inhalation was found insignificant, the thyroid equivalent dose from the internal exposure of ^{129}I using a value of 10^{-8} for the isotopic ratio ^{129}I/^{127}I in the thyroid and ICRP reference man was calculated. The corresponding thyroid cancer risk factor of 10^{-11} for an adult from life-time exposure is one order of magnitude higher than for a 1-year old child. Due to low radiation toxicity of ^{129}I the annual dose is 8×10^{4} times lower than the dose limit of the National Research Council, USA which is 0.04 mSv y^{-1} to whole body or any organ for a combined beta and photon emitting radionuclide.

1. Introduction

Iodine is an essential trace element for humans. As a biophilic element, it is mainly incorporated into the thyroid hormones. Iodine is thus required for proper functioning of the thyroid gland (recommended uptake of stable iodine through inhalation is 0.29 μg/d and by ingestion is 220 μg/d as given in UNSCEAR, 2000) and its deficiency can lead to severe metabolic disorders. Like stable iodine, radioactive isotopes are concentrated in the human thyroid (VanMiddlesworth et al., 2000), thus, uncontrolled release of these isotopes would constitute a direct threat to human populations (Hu et al., 2009). While in case of an accident short-lived isotopes are removed from the environment quickly, the introduction of ^{129}I with its half-life of 16 million years is cumulative in the environment from the standpoint of assessing human exposures.

Among 25 radioactive isotopes of iodine, the long half-life, high mobility and risks coupled with its growing inventory in the environment makes ^{129}I problematic in the long term (Zhang et al., 2010). Before the nuclear era, the natural atom ratio of ^{129}I/^{127}I was in spatial and temporal equilibrium at $(0.04–3) \times 10^{-12}$ (Michel et al., 2012). The ^{129}I releases from nuclear weapon testing during 1950–1960's leading to (^{129}I/^{127}I $= 10^{-11}$–10^{-9}), the Chernobyl accident (ratios of 10^{-9}–10^{-6} in environmental samples from the contaminated area) and especially discharges from European reprocessing plants (10^{-6}–10^{-4} near reprocessing plants) have overwhelmed its natural abundance (Hou et al., 2009a and references therein). The major pathway of anthropogenic ^{129}I in the environment is releases from reprocessing plants. For Sellafield (GB) 85% and La Hague (F) 97% of all discharges are in liquid form into Irish Sea and English Channel respectively (corresponding to ratios of 10^{-6}), while the reprocessing plant at Marcoule (F) emitted about 76% of ^{129}I in gaseous form (Michel et al., 2012; Hou et al., 2009a). The amount of gaseous ^{129}I releases from the French reprocessing facilities dropped in the late 1990s (Marcoule was decommissioned in 1997), so that the main source of gaseous ^{129}I releases is now located in Great Britain (Reithmeier et al., 2005, 2010). The main form of released iodine from these sources to the atmosphere is alkyl halide (Buraglio et al., 2001; Persson et al., 2007). Within the troposphere, long-ranging transport of the organic gaseous form is likely, given the estimated residence time of 18 days (Moran et al., 1999). These iodine compounds can be photo-dissociated in the atmosphere and subsequently oxidised to water soluble iodine species (iodate and iodide) and adsorbed to the aerosols (Buraglio, 2000). But there is a discrepancy found between most model calculations, which suggest that iodate should be the dominant iodine species in the particulate phase

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(Vogt et al., 1999; McFiggans et al., 2000) and field observations which show that organically bound iodine and iodide are the dominant iodine species (Baker et al., 2001; Baker, 2005; Gilfedder et al., 2007). Depending on the residence time of iodine species, the atmospheric discharged ^{129}I is transported on global scale and distributed from the Northern to Southern hemisphere (Suzuki et al., 2010). Marine discharged ^{129}I is used as oceanographic tracer (Synder et al., 2010).

Although the ^{129}I level has increased considerably it is radiologically not hazardous since the concentration required to deliver significant dose is relatively high due to its low specific activity (6.55 MBq g^{-1} UNSCEAR, 2000). However, ^{129}I is one of key risk drivers at nuclear waste repositories due to uncertainty regarding its biogeochemical fate and transport in the environment (Denham et al., 2009); therefore investigation about its behaviour has become indispensable for planning of future environmental assessment and mitigation efforts. In the UNSCEAR (2000), ^{129}I was identified as a globally dispersed radionuclide.

Despite increasing concentrations of ^{129}I in the environment, which today are up to 10^6 times the prenuclear values in Northern Europe (Englund et al., 2008), only a few studies concerning the distribution of ^{129}I in the atmosphere as short time series have been reported (Aldahan et al., 2009; Englund et al., 2008; Hou et al., 2009b; Lopez-Guiterrez et al., 2000, 2004; Michel et al., 2012; Santos et al., 2006; Reithmeier et al., 2006; Szidat, 2000). Among them only one report of a retrospective study of ^{129}I in air exists (Englund et al., 2010). Consequently, up to now little is known about pathways of ^{129}I and many questions concerning the influence of meteorological conditions, locations of deposition and percentage contribution of gaseous or liquid releases from the European reprocessing plants are still open. To elucidate this problem at least partially, we conducted a study about temporal distribution of ^{129}I in air in the course of year 2001 and concluded that most ^{129}I in air particles basically originated from gaseous emissions from the Sellafield reprocessing plant (Jabbar et al., 2011). This has intrigued us to extend the 2001 study to the following year 2002. As also older air filters were available at the Institute of Inorganic Chemistry, we were able to reconstruct the long-term atmospheric trend of ^{129}I over four decades back to the 1960s. From this data, potential radiological impacts of ^{129}I on general public and the possible health detriment in terms of cancer risk were estimated.

2. Materials and methods

Airborne particles (aerodynamic diameter 1.6 μm) were collected at meteorological stations Hohe Warte (48°15′ N and 16°21′50″ E; 202 m above sea level) during 1960s to 1980s and at Retz (48°45′39″ N and 15°56′30″ E; 323 m a.s.l.) in the 1990s. Aerosol filter samples stored in a dark and cool place were combined to make yearly representatives from selective years during 1966–2002.

To check temporal variation of ^{129}I, aerosol samples were taken weekly (Hohe Warte) during 2001–2002 at the height of 1.2 m above ground level with a flow rate of around 80 m^3/hour air using 20.3×25.4 cm^2 glass microfiber filters. Weekly samples of the year 2001 were combined to biweekly samples (Jabbar et al., 2011); for the present work on the year 2002, however, weekly samples were processed to have better resolution of short-term fluctuations corresponding to anthropogenic releases. In a few weeks after sampling, the air filters were first measured by a REGe Detector (Canberra GR 2020) to determine ^{7}Be concentrations. The counting time of weekly collected samples was 1000 min. For the determination of the ^{7}Be concentration the photopeak at 477.6 keV was used.

The radiochemical method of ^{129}I has been described somewhere else (Jabbar et al., 2011) so only a brief description is given here. Iodine was extracted from the filter paper in a strong alkaline solution of 0.5 M NaOH and 0.05 M $NaHSO_3$. After filtration through a 0.45 μm cellulose acetate membrane filter, all the iodine in solution was reduced to iodide at pH <2. Stable iodine concentration was measured on an aliquot by ICP-MS Agilent 7500ce, Waldbronn, Germany, equipped with a CETAC ASX-520 autosampler (using Cs as an internal standard in 2% ammonia solution) before adding potassium iodide carrier (5 mg of I^-). Since carbon tetrachloride which is traditionally used to extract iodine from an aqueous phase has significant environmental, health, and regulatory problems associated with its use, we performed experiments to find an alternative system. Pre-conditioned 10 g of Dowex 1×8 (100–200 mesh Cl^- form) was used to separate iodine from matrix elements. Iodide was eluted with 200 mL of 0.5 M HNO_3. The chemical yield of the procedure, determined by ICP-MS, ranged from 70 to 85%. Subsequently, iodine was precipitated as AgI by addition of $AgNO_3$. The precipitates were washed with 25% ammonia and 3 M nitric acid followed by washing with millipore. Finally, precipitates were dried at 60 °C, mixed with silver powder (AgI:Ag 1:1 by weight) and pressed into copper targets for Accelerator Mass Spectrometry (AMS) measurement. Eventual contamination introduced during the process was determined by the preparation of blank samples processed in the same way as real samples.

AMS measurements were carried out at the Vienna Environmental Research Accelerator (VERA). The $^{129}I/^{127}I$ ratio was determined using 3 MV terminal voltage followed by selection of charge state +4 (Wallner et al., 2007). Ratios were normalized using a standard material "ETH-D2" provided by the ETH Zürich with a nominal isotopic ratio of 4.73×10^{-11}. This material was produced by dilution of the NIST reference material SRM 4949B-79 (Synal, 1998). The typical $^{129}I/^{127}I$ ratio of the blanks was 10^{-13} and the samples showed $^{129}I/^{127}I$ ratios of 10^{-11}–10^{-9}. The measurement uncertainty was <5% (one standard deviation).

3. Results and discussion

Airborne particles were analysed by ICP-MS and AMS to determine ^{127}I and ^{129}I/I isotopic ratio, respectively. The concentration of ^{129}I was derived from the isotopic ratio using the concentration of stable iodine and chemical yield for iodine separation determined from ICP-MS. Only the chemical yield of the first step, namely leaching of air filters by reflux heating in alkaline medium could not be controlled. According to Hou et al., loss of iodine at pH 9–10 and 60–80 °C is negligible (Hou et al., 1998).

3.1. Temporal variability of iodine-129 during 2001–2002

The overall range of stable iodine over the two years was 0.2–2.96 ng Nm^{-3}, generally less in summer compared to winter and at the lower end of the 1—100 ng Nm^{-3} range reported in the literature (Qinhong and Moran, 2010). The trend of relatively lower iodine concentration may be partly related to distance from the coast, the type and concentration of aerosols in the atmosphere, the sources of air masses, dust mass particle and the seasonal variation of dust mass particle type (Buraglio et al., 2001; Englund et al., 2010). The $^{129}I/^{127}I$ isotopic ratio showed great variation from 0.6×10^{-8} to a maximum of 45.8×10^{-8}. The frequency distribution of ^{129}I concentration measured in aerosols during 2001–2002 is shown in Fig. 1. The solid curve shows a fit to the data with a skew normal distribution of skewness 1.1. Using the Student's *t*-distribution, the 95% confidence interval for ^{129}I concentration (A.M. $= 4 \times 10^5$ atoms Nm^{-3}) in air over a period of two years was

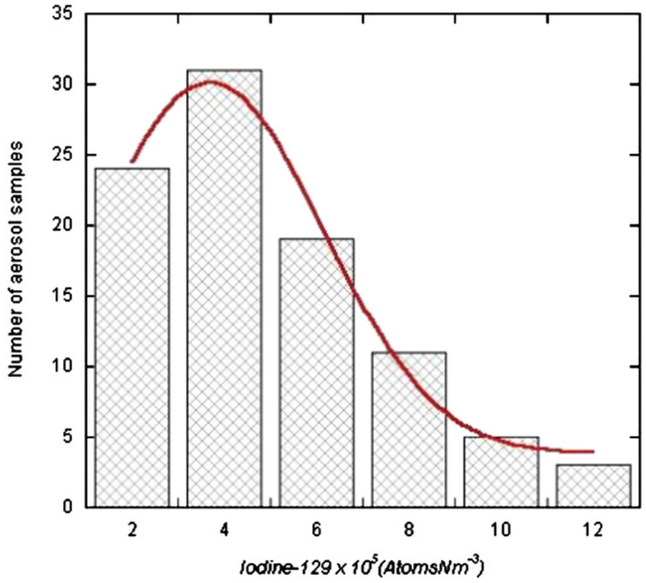

Fig. 1. Frequency distribution of ^{129}I concentration for the year 2001–02.

found to be (4.57–3.38) × 10^5. The wide range of variation in the ^{129}I shows that major sources and pathways of ^{129}I are rather different from ^{127}I. Gaseous releases and the fraction of liquid releases volatilized from the ocean surface have been shown to be the main source of ^{129}I deposited over Europe (Aldahan et al., 2009; Hou et al., 2009a, 2009b; Keogh et al., 2010). However, there is a controversy about possible contribution of atmospheric ^{129}I from sea spray (Baker, 2004; Moran et al., 1999; Reithmeier et al., 2006, 2010). To negotiate this hypothesis, we compared the time distribution of ^{129}I concentration to temporal evolution of gaseous ^{129}I emitted by Sellafield and La Hague (Fig. 2).

Our data clearly indicate strong temporal fluctuations originating from direct airborne ^{129}I releases. Comparable to previous findings during 2001, ^{129}I air concentration followed the pattern of gaseous emission from Sellafield. Peak values of ^{129}I were also supported by weather data with predominant wind direction from west and north-west during sampling time (Jabbar et al., 2011). However, we found deviations at certain points. Since only data on monthly ^{129}I releases were available, it may have happened that emissions from La Hague and Sellafield from end of February were reflected completely or partially in the aerosol sample corresponding to March 2002. Velocity and pattern of wind over Western Europe and precipitation may also influence the distribution of ^{129}I in the atmosphere. Another parameter that might modulate the level of ^{129}I is the vertical air mass transfer. As ^{7}Be is considered a good tracer to study stratosphere-to-troposphere exchange and downward transfer, we compared monthly ^{129}I concentrations with ^{7}Be activities (Fig. 3) (Jabbar et al., 2011). Elevated levels of ^{7}Be seem to be supported by vertical air transport that suppresses ^{129}I concentration.

3.2. Retrospective time profile of iodine-129

The ^{129}I concentration in aerosol samples from Austria covering the period 1966–2002 was measured (Fig. 4). While fission products from atmospheric atomic bomb testing show a clear decrease after the 1963 test ban treaty, here a gradual increase from 1965 till 1984 is visible followed by a decrease from 1984 onward, similar to the pattern of gaseous discharges from European fuel reprocessing plants. This is in accordance with Reithmeier et al. (2010) who also observed that ^{129}I signals produced by the nuclear weapon tests during 1950–1960's (and by the Chernobyl accident in 1986) were overwhelmed by the contribution from the reprocessing plants. During 1977 to 1995, the NRP at Marcoule (in Southern France) released ^{129}I mainly to the atmosphere at a relatively high rate (7–10 kg y^{-1}) compared to Sellafield (2–6 kg y^{-1}) and La Hague (2–6 kg y^{-1}) and contributed considerably to the peak values of ^{129}I (Marcoule was decommissioned in 1997). The relative contribution of ^{129}I releases from different sources strongly depends on the sampling region. Contrary to our findings, the aerosol ^{129}I time series from Southern and Northern Sweden showed a gradual increase during 1985–2005 (Englund et al., 2010). In this case the experimental data indicated the dominating impact from liquid emissions (which became airborne by sea spray) typical for near-

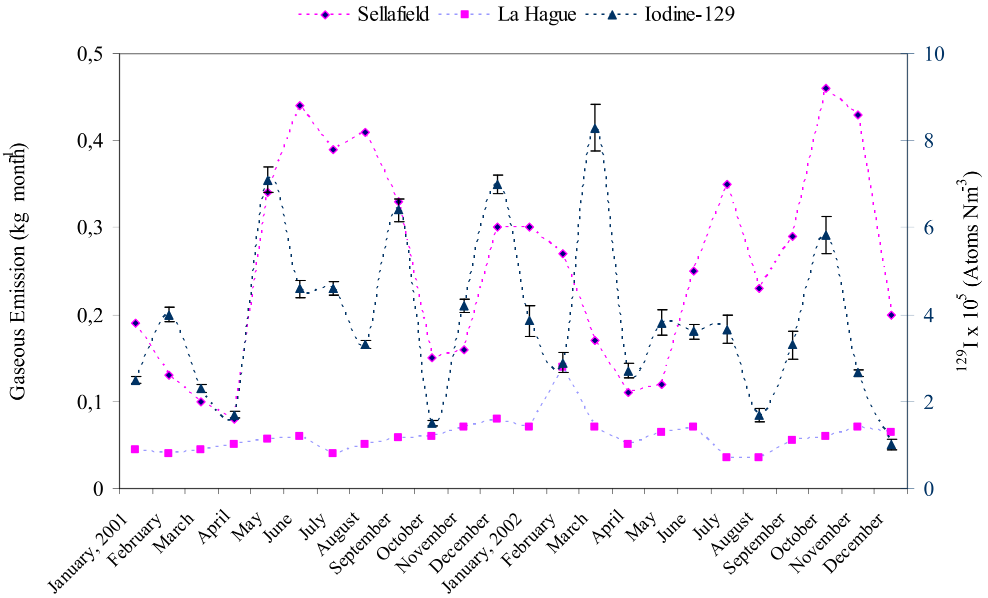

Fig. 2. Temporal variation of ^{129}I concentration in aerosols from Vienna, Austria compared to monthly gaseous emissions of ^{129}I from the reprocessing facilities at La Hague and Sellafield. ^{129}I data of the year 2001 was taken from Jabbar et al. (2011).

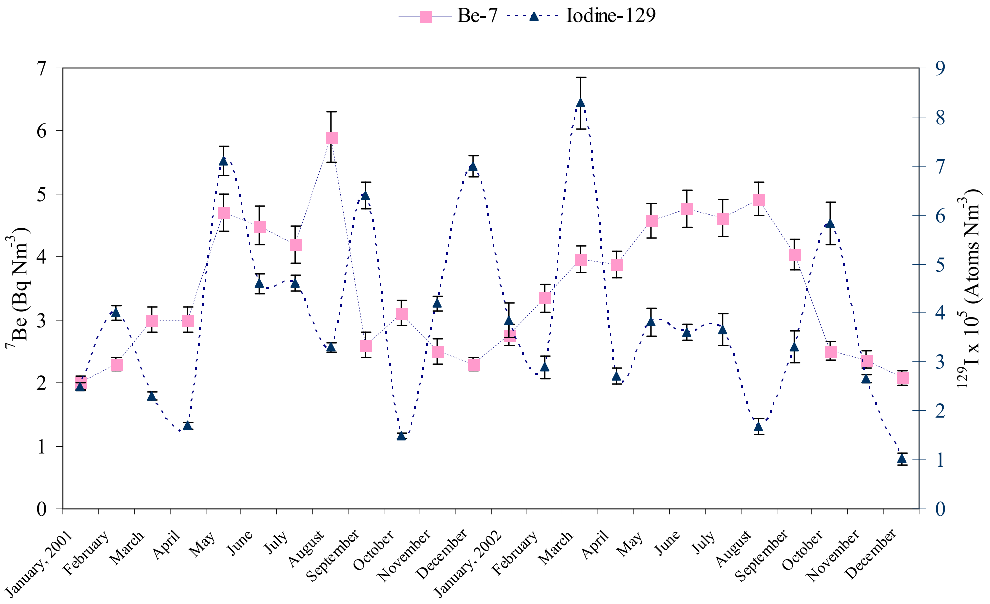

Fig. 3. Comparison of monthly ^{129}I concentrations with ^{7}Be activities.

coastal sites. For inland areas like Vienna, however, liquid releases are insignificant compared to gaseous emissions.

Aerosols can reach ground through particle sedimentation (dry deposition) and more efficiently by precipitation scavenging (wet deposition), both depending on the associated particle size distribution. It is a major source of contamination of terrestrial food products and pasture vegetation. Using a deposition velocity range v_d of 0.001–0.02 m s^{-1} for iodine isotopes and the annual concentration of the respective isotope $C(t)$, the dry fallout $F_d(t)$ (given in atoms cm^{-2} y^{-1}) is estimated by Englund et al. (2010) as

$$F_d(t) = C(t)^{*}v_d^{*}\mathrm{tr}$$

(tr = 3155.76 is the conversion factor from m^{-2} s^{-1} to cm^{-2} y^{-1}). Values of dry fallout calculated from ^{129}I concentrations are given in Table 1. Unfortunately we do not have data of ^{129}I concentration in precipitation, and therefore we cannot constitute the absolute percentage of dry and wet deposition. However, Englund et al. concluded that dry fallout (constitutes 1/4 of total fallout at the southern Sweden and 1/3 at the northern site) is of minor importance in comparison with wet deposition.

3.3. Iodine-129 dose assessment

The contribution of ^{129}I to inhalation dose was estimated using observed concentrations of this radioisotope in aerosols during 2001–02. The potential risks of internal exposure to ^{129}I are a result from both inhalation and ingestion of contaminated food, however, the amount of iodine ingested by far exceeds the amount inhaled.

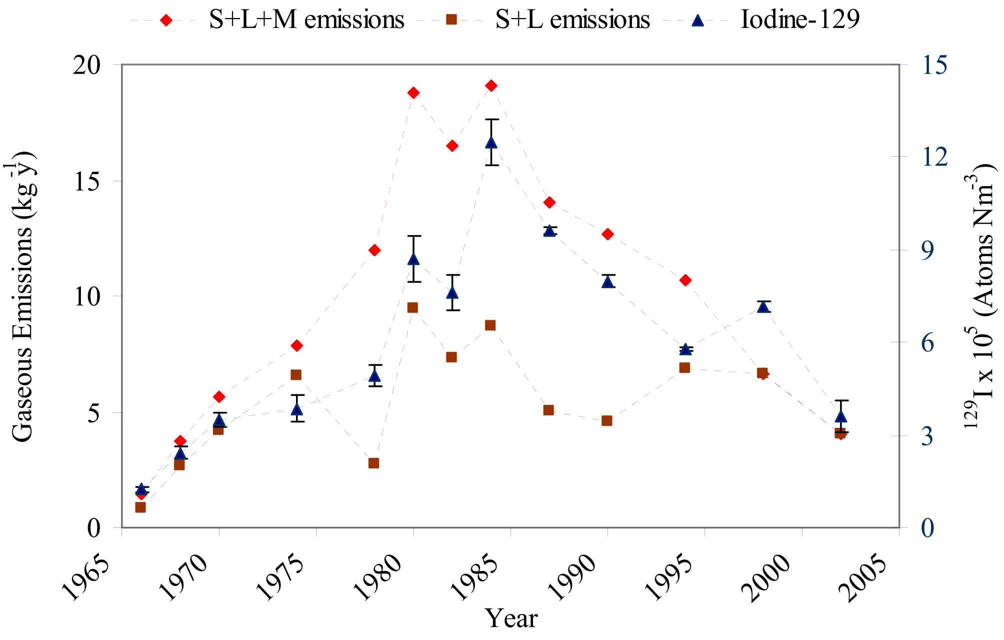

Fig. 4. Time profile of ^{129}I concentration and total gaseous ^{129}I emissions from European NRP (Sellafield, La Hague and Marcoule) over 4 decades.

Table 1
Dry deposition of iodine isotopes at Vienna using range of deposition velocity of 0.001–0.02 m s^{-1}.

Year	^{129}I (10^5 atoms m^{-3})	^{129}I (10^5 atoms $(cm^2y)^{-1}$)	^{127}I (ng m^{-3})	^{127}I (ng $(cm^2y)^{-1}$)
1970	3.5	13–220	3.5	11–227
1980	8.7	27–547	2.9	9–184
1990	7.9	25–503	2	5–106
2001	4.2	11–227	0.9	2–57

The same is true for ^{129}I, as the anthropogenic ^{129}I has been dispersed globally. The radiological impact of a given level of ^{129}I activity depends on the stable iodine uptake, the degree to which the thyroid of individuals is saturated with stable iodine, on the thyroid mass and on the ^{129}I concentration in the air or foodstuffs (Guen et al., 2000). The radiation dose from ^{129}I is almost entirely due to a low energy beta particle that has a very short range in tissue. Iodine-129 inhaled or ingested is transferred into the blood with most of iodine uptake concentrated into the thyroid gland (with the biological half-life of 20–100 days for 1 year old child and an adult, respectively, as reported by Soldat, 1976). As a result tissues that receive the highest radiation doses are the thyroid gland and the surrounding tissues (e.g., parathyroid gland). High concentration of radioiodine in the thyroid gland significantly increases the risk of thyroid cancer development.

To determine radiological impact, equivalent dose from inhalation of ^{129}I as given by Avila and Facilia (2006) and Syrakov et al. (2009);

$$D_{\mathrm{inh}}^{\mathrm{I-129}} = C_{\mathrm{air}}^{\mathrm{I-129}} * \mathrm{InhR} * H * \mathrm{DC}_{\mathrm{inh}}^{\mathrm{I-129}} \tag{1}$$

Where $C_{\mathrm{air}}^{\mathrm{I-129}}$, InhR and H denote concentration of ^{129}I in air (Bq m^{-3}), the inhalation rate (m^3 air day^{-1}) of an individual and 365 d y^{-1} exposure time to contaminated air respectively, and $\mathrm{DC}_{\mathrm{inh}}^{\mathrm{I-129}}$ denotes dose coefficient for inhalation. The relevant data for exposure due to inhalation pathways are listed in Table 2 (ICRP, 1995).

Applying annual inhaled air by 1-year old child and an adult (assuming ICRP, 1975 intake of 5.1 and 22.2 m^3 air day^{-1}) and average ^{129}I concentration in air (5.64×10^{-10} Bq Nm^{-3}), total intake of airborne ^{129}I is 1 and 4.5 μBq y^{-1}, respectively. This intake is negligible considering EPA derived NESHAPs (National Emissions Standard for Hazardous Air Pollutants) of 0.29 Bq m^{-3} in air (Human Health Fact Sheet, 2007). The equivalent dose to the thyroid gland is derived to be 10^{-9} mSv y^{-1} as given in Table 2. One must take into account that only ^{129}I associated with aerosols is considered for estimation of inhalation doses. Since iodine exists as organic, inorganic and particulate in the atmosphere, the relative contribution of different species is wide and uncertain. Measurement of ^{129}I species made by Michel et al. (2005), in the air over the North sea indicates particle associated, inorganic and organic gaseous were in the range of 18%, 43% and 40% respectively with similar distribution for ^{127}I. Considering these results, inhalation dose values comprising all iodine species might be higher by a factor of 6.

Table 2
Doses from internal exposure of iodine-129 inhaled.

General public	Annual inhaled air (m^3 y^{-1})	Dose coefficient thyroid (Sv Bq^{-1})	Equivalent dose thyroid 10^{-12} (Sv)
1-year old child	1860	1.7E-06	1.7
Adult	8100	7.1E-07	3.2

However, as already mentioned, the amount of ^{129}I inhaled is insignificant compared to the amount taken up via ingestion. The total annual dose equivalent to the thyroid is calculated from the total amount of ^{129}I in this organ. The value was found to be on the order of $1.1–5 \times 10^{-7}$ mSv y^{-1} using the isotopic ratio $^{129}I/^{127}I$ of 10^{-8} in the thyroid (Handl et al., 1993), ICRP reference man for human iodine content: 1 mg stable iodine in 1 year old child and 10 mg stable iodine in adult thyroid and dose coefficient as given in Table 2. This derived value is approximately 8×10^4 times lower than the National Research Council USA dose limit of 0.04 mSv y^{-1} for combined beta and photon emitting radionuclides to whole body or any organ (Hou et al., 2009a).

Using ICRP fatal thyroid cancer probability coefficient of 8×10^{-4} Sv^{-1}, and life-time (70 and 50 years for 1-year old child and adult) internal exposure of 10^{-9} and 10^{-8} Sv, the associated life-time cancer risk is estimated as (ICRP, 1991; Jabbar et al., 2009),

$$\mathrm{Risk} = \mathrm{Dose(Sv)} * \mathrm{Risk\ coefficient}\left(\mathrm{Sv}^{-1}\right) \tag{2}$$

Equation (2) gives a life-time cancer mortality risk of 10^{-11} for an adult which is one order of magnitude higher than for a 1-year old child. Again this value is a negligible fraction of the total risk (6×10^{-3}) from all natural radiation sources based on global average annual radiation dose of 2.4 mSv yr^{-1} to man (Akhter et al., 2007).

4. Conclusions

Our study provides the time profile of ^{129}I in air for a European region not directly affected by local releases. The general trend of ^{129}I in the air particles during 1966–2002 suggests a dominant impact from atmospheric emissions by three European reprocessing plants. Monthly temporal fluctuations in 2001 and 2002 were governed by gaseous emissions from Sellafield and meteorological conditions. The radiological impact of ^{129}I was found insignificant in comparison to the EPA derived National Emission Standard for Hazardous Air Pollutants of 0.29 Bq m^{-3} for ^{129}I in air. Thus our study concludes that presently ^{129}I is not a radiological hazard but due to its long half-life and continuous emission from reprocessing plants could become significant from radiation protection point of view and therefore should be monitored continuously.

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