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Iodine-129 in animal thyroids from Argentina

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ABSTRACT

^{129}I and ^{127}I concentrations in animal thyroids coming from several regions of Argentina were determined by accelerator mass spectrometry (AMS) and gas chromatography (GC), respectively. The measured ^{129}I/^{127}I ratios, ranging from 3×10^{-12} to 4×10^{-10}, are significantly lower than those typical for areas in the northern hemisphere (10^{-10}–10^{-7}). The ^{129}I concentrations show a clear dependence with latitude and season, which can be understood considering tropospheric circulation patterns, possible ^{129}I sources and regional precipitation rates.

1. Introduction

^{129}I is a long-lived ($T_{½}=15.7$ Myr) radioisotope which is naturally formed either from fission of uranium in the Earth's crust or from the interaction of cosmic rays with atmospheric xenon isotopes. Accounting for these processes, the natural world inventory of ^{129}I has been estimated to be around 50,000 kg (Fabryka-Martin et al., 1985), from which only approximately 140 kg corresponds to the hydrospheric inventory (Snyder et al., 2010). Since the beginning of the nuclear era, the amount of ^{129}I has increased by 45 kg due to nuclear weapon tests and nuclear accidents (Schmidt et al., 1998) and since the late 1960s surface inventories have further increased due to nuclear fuel reprocessing plants in the northern hemisphere, whose activities have added almost 5400 kg (Snyder et al., 2010). Still, in the Southern hemisphere the main contribution to the deposition of ^{129}I is the fallout from atmospheric nuclear explosions (Reithmeier et al., 2010; Snyder et al., 2010). At present in the austral hemisphere there are no declared reprocessing plants and only four nuclear power plants are operative, two of which are located in Argentina (with another under construction).

The present work aims to investigate the presence of ^{129}I in the Southern hemisphere by determining ^{129}I/^{127}I ratios in the thyroids of grazing animals at different latitudes within Argentina. Cattle generally contain 10 to 30 mg of iodine in their bodies, of which more than 90% is concentrated in their thyroid glands (NCRP, 1983). Thyroid hormones require iodine for their synthesis and therefore this element is naturally concentrated in thyroid glands, up to 5 orders of magnitude greater than typical concentrations in pasture and other types of cattle feed. Iodine content in thyroids is modulated by its biological half-life, which is estimated to be 100 days, and provides a time-integrated signal (NCRP, 1983). The iodine content is also affected by seasonal variations due to changes in animal metabolism and iodine content in the animal's diet (Miller et al., 1975).

The major intake of iodine in cattle is via the ingestion of fresh pasture. Consequently, thyroids of grazing animals act as sensitive probes to assess the fallout of ^{129}I in environmental studies, such as retrospective dosimetry of the ^{131}I exposure after the Chernobyl accident (van Middlesworth et al., 2000). Optimally, this type of study would require background values; yet, prior to this work, there is no ^{129}I data from cattle-thyroids in Argentina.

In this work, iodine-129 and iodine-127 concentrations from animal thyroid samples were measured by accelerator mass spectrometry (AMS) and gas chromatography (GC), respectively. To our knowledge, these are the first AMS measurements of thyroid samples obtained in the Southern hemisphere. In addition, our study covers a large latitudinal range allowing us to interpret the latitudinal distribution of ^{127}I and ^{129}I and we speculate about their respective behavior in the environment.

2. Experimental

Between the Winter 2009 and the Summer 2010, fresh thyroids of grazing animals (one lobe per animal, not coming from feedlots) were collected from slaughter houses in ten different locations (See the locations in Fig. 1). Animals with an age of 3 to 4 years were

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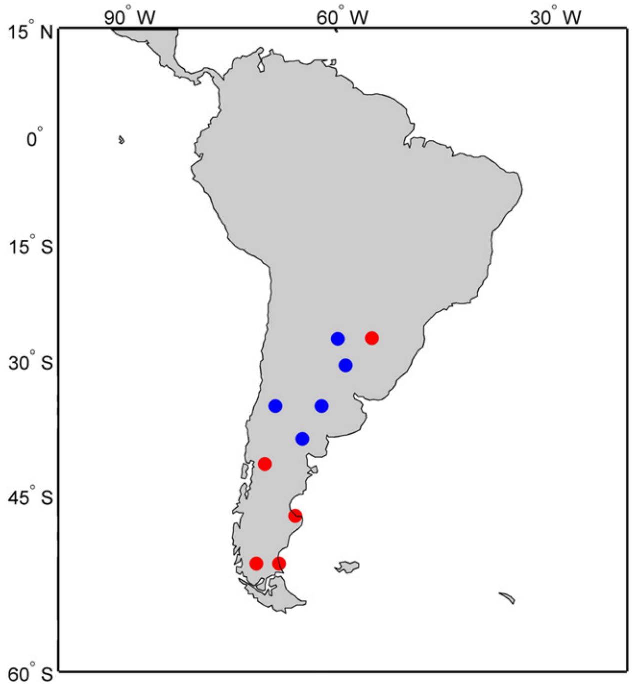

Fig. 1. Map of South America showing the sampling point locations. According to their collection time, samples were divided in two sets (April — red circles, Winter — blue circles). See Table 1 for details.

taken from a herd that grazed fresh pastures at known locations close to the slaughter houses for at least one year prior to their thyroid donation. The studied animals did not receive any dietary supplement; therefore, it is assumed that there is no difference in the diet of the studied animals. Animals from the same herd were of the same breed. The thyroids of one to five cattle were analyzed form each sampling site (See Table 1). The samples were weighed and then frozen for further preparation in the laboratory.

After being lyophilized and homogenized they were chemically processed at the TANDAR Laboratory in Buenos Aires. Extraction of iodine was based on two methods developed at LLNL (Marchetti et al., 1997; Gu et al., 1997), adapted for this particular matrix and the very low ^{129}I concentrations expected in these samples.

We selected 100 mg of dry-freezed thyroid and applied the following procedure: first, the sample was oxidized in a solution of peroxydisulfate (Gu et al., 1997). From this solution, we prepared the derivative to determine total iodine by gas chromatography (GC) and the samples ready for AMS measurement. In a separation funnel, we combined 200 µl aliquot of the sample solution with 25 ml of water, 1 ml of 4% pentan-3-one, 1 ml of 30% hydrogen peroxide and 2 ml of 5 M sulphuric acid. The derivative was extracted in hexane. The recovery for the extraction method for GC was 75–80%, estimated using a NIST standard reference material (SRM-1549), non-fat milk powder with a reference value of (3.38 ± 0.02) µg/g. For the determination of ^{129}I, 1.0 g of Na_2SO_3 was added to the rest of the sample solution to reduce iodate to iodide. Within a separation funnel, 2 ml of a KI solution (Merck Suprapur®, 1000 µg/ml), acting as carrier was added to the solution sample, followed by concentrated nitric acid to adjust pH ~ 1 and then 400 µl of silver nitrate solution (5% m/V), to precipitate silver iodide. The precipitate was thereafter washed and left in the oven to dry overnight and finally mixed with silver and pressed in a sample holder for accelerator mass spectrometry (AMS) measurements.

^{129}I/^{127}I ratios of the prepared samples were measured at the Vienna Environmental Research Accelerator (VERA) (Steier et al., 2005). In this facility, an I^- ion beam is generated in a Caesium sputter ion source. The masses 127 and 129 are alternatively selected and injected into the tandem accelerator by adjusting their particle energy in the chamber of the injection magnet. The timing of such a switching cycle was generally 0.2 s with ^{129}I being injected most of the time. The negative ion beams are accelerated by the tandem accelerator which is held at a voltage of +3 MV. In the terminal of the accelerator, the particles pass a gas section where the negative ions lose electrons in a stripping process. Positively charged ions are then accelerated again from the positive terminal. After passing the tandem, the positively charged particles are filtered by a Wien filter, a 90° analyzing magnet (where the charge state 4+ was actually selected), an electrostatic analyzer and a switching magnet. While the $^{127}I^{4+}$ beam is measured with a Faraday cup (currents between 0.1 and 3 µA), the $^{129}I^{4+}$ ions are identified in the detection system consisting of a Time-of-Flight setup followed by a Bragg particle detector.

In order to estimate possible contributions of ^{129}I due to interfering events in the detector and/or contamination from the chemical reagents, machine- and chemistry-blanks were prepared, respectively. Ratios as low as $(8.3 \pm 2.4) \times 10^{-14}$ were measured for machine-blank samples (samples with low ^{129}I concentration expected). Chemical-blanks, prepared following the above mentioned chemistry steps but without adding thyroid samples, allow to evaluate the ^{129}I level of the reagents. The background estimated from chemical- and machine-blank values were subtracted from the measured ^{129}I/^{127}I sample ratio. Samples were at least one order of magnitude above the chemical-blank level, $(3.6 \pm 0.5) \times 10^{-13}$,

Table 1
^{127}I, ^{129}I contents and ^{129}I/^{127}I ratios measured in animal thyroid samples of several locations of Argentina. Dry wt means dry weight of thyroid.

Sample #	Location	Date	Number of samples (n)	Iodine [mg/g dry wt.]	Atoms ^{129}I/g dry wt. (10^8)	^{129}I/^{127}I (10^{-11})	Lat (S)	Long (W)
1	Villa Ángela	Jul-09	5	5.3 ± 0.3	14.5 ± 0.7	5.7 ± 0.4	27.5	60.7
2	La Paz	Jul-09	5	3.0 ± 0.3	2.8 ± 0.4	2.0 ± 0.3	30.7	59.6
3	Rivadavia	Jul-09	5	3.1 ± 0.3	1.9 ± 0.1	1.2 ± 0.1	35.5	63
4	Malargüe	Sep-09	5	2.4 ± 0.3	2.6 ± 0.2	2.3 ± 0.2	35.5	69.5
5	Choele Choel	Sep-09	5	0.5 ± 0.3	2.1 ± 0.2	8.2 ± 0.8	39.2	65.7
6	Posadas	Apr-10	3	4.6 ± 0.3	84 ± 2	38 ± 2	27.4	55.9
7	Ñorquinco	Apr-10	3	1.6 ± 0.3	12.3 ± 0.5	16 ± 1	41.9	71
8	B. Mazaredo	Apr-10	3	8.1 ± 0.3	4.4 ± 0.4	1.1 ± 0.1	47.1	66.7
9	Río Turbio	Apr-10	3	5.8 ± 0.3	1.7 ± 0.2	0.6 ± 0.1	51.5	72.2
10	Río Gallegos[a]	Apr-10	1	6.2 ± 0.3	0.9 ± 0.1	0.3 ± 0.1	51.5	69

[a] Ovine thyroid.

except for the Rio Gallegos sample which was just a factor 3 larger than the blank.

The AMS values obtained for the ^{129}I/^{127}I concentration ratio of thyroid samples were obtained after normalization to a standard material. This material was measured along with thyroid samples for a periodical determination of the accelerator transmission and of the detection system efficiency for the counting of ^{129}I events.

3. Results

In the following, the analysis of the data was done assuming that no correlation exists between the iodine content in soils and plants growing on them. Actually, the root uptake of iodine is relatively unimportant for the iodine content of the edible part of the plant being the most significant pathway the direct absorption from the atmosphere (Fuge, 2005; Whitehead, 1984).

3.1. The ^{127}I isotope

The ^{127}I content of the analyzed thyroids ranges between 0.5 and 8.1 mg/g (dry weight), which agrees with typical values in previous studies (Chao and Tseng, 1996). The obtained iodine concentrations as a function of latitude are shown in Fig. 2. The iodine content decreases from north to South until around 40°S and further southward, higher iodine values are observed once again. This particular latitudinal profile can be explained with local tropospheric circulation patterns (See Fig. 3) and considering the Ocean as the principle source of atmospheric iodine — from which iodine is transferred to the atmosphere and washed out by precipitation to the marine and terrestrial environments (Moran et al., 1999; Whitehead, 1984).

The west branch of the Atlantic anticyclonic circulation delivers the major contribution of moist air in northern and central Argentina, being this input larger during the austral Summer (Vera et al., 2006). This contribution introduces humidity from the Atlantic Ocean to South-central Brazil, which is then channeled to the Andes and enters to Argentina at the east of the mountains. Wet deposition of iodine is modulated by local precipitation, which is higher in the Summer time and decline to Southern Argentina (See Fig. 3). South of latitude 40°–45°S, the air circulation pattern is quite different due to the low altitude of the mountains and the location itself between the Atlantic and Pacific anticyclone systems. As a result there is an intense western wind which brings Pacific air masses that even could interact with sporadic and local incursions of Atlantic air.

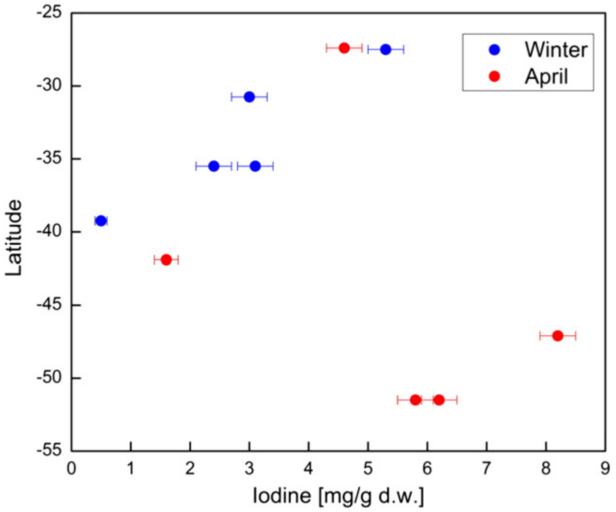

Fig. 2. Variation of stable iodine as a function of latitude (April — red circles, Winter — blue circles).

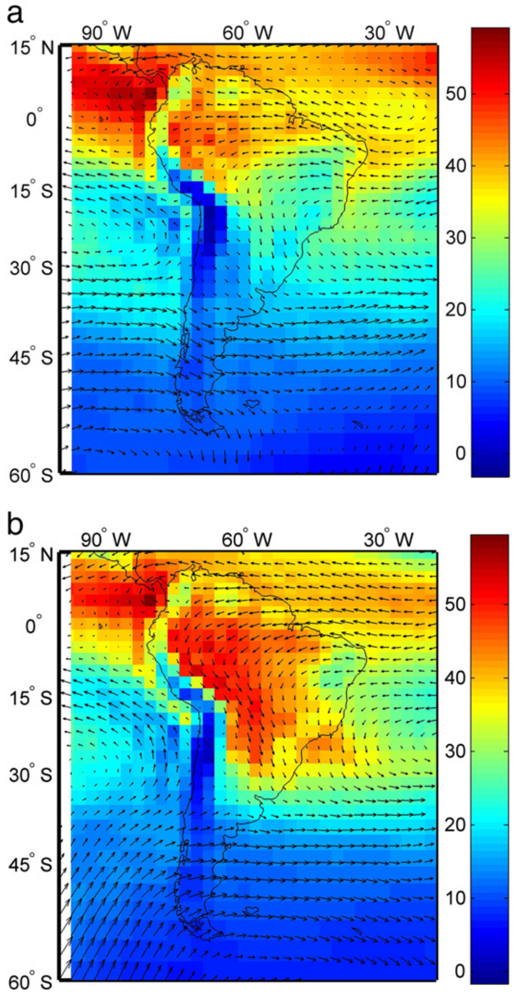

Fig. 3. Mean wind shear in m/s at 1500 m and mean precipitable water in kg/m^2 at August (a) and November (b) of 2009.

3.2. The ^{129}I radioisotope

Thyroid concentrations of ^{129}I are plotted as function of the latitude in Fig. 4. Contrarily to the pattern of the stable ^{127}I, the latitudinal distribution of the ^{129}I concentration can be classified into two distinguishable groups. These groups were labeled according to their collection time, samples 1 through 5, taken during the austral Winter season (July–September), and samples 6 through 10 taken during April (See Table 1).

The distinction between Winter and April ^{129}I values suggests the presence of an active ^{129}I source. We interpret this as a recycling process rather than the introduction of new ^{129}I into the environment. The monotonic decrease of the ^{129}I concentrations indicates a potential source located to the North. This trend is also reflected in the very low ^{129}I values measured in the samples collected in Southern Patagonia, indicating that the possible source has an almost negligible contribution to this region.

Although the emission of iodine from the Ocean surface (210 Gg for methyl iodide/year) was already identified as a major contributor in marine and terrestrial environments, biomass burning is another known iodine source (3–10 Gg for CH_3I/year) (Bell et al., 2002). We propose that the burning of biomass might play a significant role as ^{129}I recycling source in South America. It is well known that accidental fires and intentional burning of firewood mobilize radionuclides

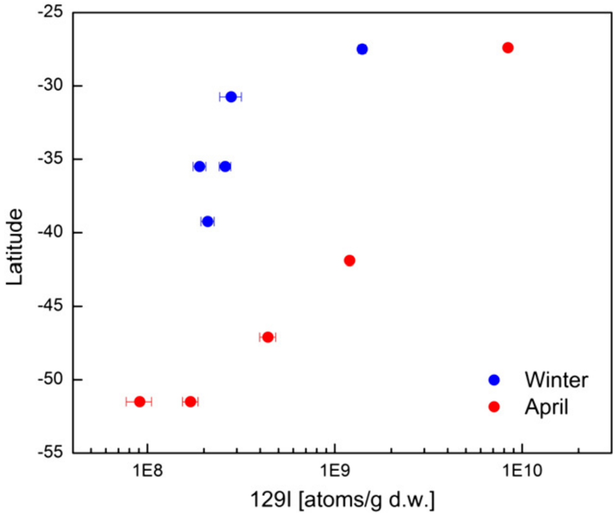

Fig 4. Variation of ^{129}I as a function of latitude (April — red circles, Winter — blue circles).

through suspension of gasses and particles in the atmosphere. Indeed about 90% of the iodine content is lost to the atmosphere during combustion processes (Amiro et al., 1996). Considering the widespread deposition of ^{129}I across the entire surface of the Earth (Reithmeier et al., 2010; Snyder et al., 2010), the large inventory of the stable iodine in the Ocean water makes the ^{129}I/^{127}I ratios lower in marine samples than in terrestrial ones. Widespread burning, mainly tropical forest and savannah, is a common practice in South America and it is the major source of atmospheric pollution at continental scale, affecting annually around 10,000 km^2 in the Brazilian Amazon (Mongabay, 2012).

Biomass burning products reach the northern and central part of Argentina (Otero et al., 2011) carried by the humid wind which passes through the Amazonian forest. This transport is enhanced by the presence of more intense meridian flow and low-level jet events during the austral Summer (Freitas et al., 2005). Therefore, taking into account this fluctuation of the transport pattern and the seasonal dependence of the biomass burning in Brazil with higher activity during dry season (July–December), the deposition of ^{129}I in the Northern and central regions of Argentina should also present seasonal variations. In this way, although there is an almost constant annual contribution of the iodine content (of both isotopes) coming from the Atlantic (Bell et al., 2002), one might conclude that austral Summer samples have an extra contribution of ^{129}I stemming from biomass burning in Brazil. Therefore, in samples taken during the austral Winter (when both isotopes predominantly originate from the Atlantic Ocean) one should expect a large positive correlation between concentrations of the ^{127}I and ^{129}I. This is reflected in the Pearson correlation coefficient r = 0.81 obtained for the samples taken between July and September. On the other side, in burning season this correlation should be broken because of the appearance of the ^{129}I recycling phenomena.

Samples from Patagonia have to be considered in a different way. Besides a weak contribution of the Amazonian biomass burning to the ^{129}I inventory, strong winds from the Pacific Ocean are probably the main source of iodine isotopes to this region.

One way to survey the influence of the recycled ^{129}I from biomass burning is by studying the sample taken in Posadas, which is the only Northern sampling site close to the burned regions and taken in early April 2010. The 2009 burning season presented a maximum activity in the North of the Amazon between September and November. We focus on November 2009, the month of larger activity in the Northern states of Pará and Maranhão (65% of the fires in November in Brazil corresponds to these two states), where a burned surface of around 1700 km^2 was identified (CPTEC, 2011). The total ^{129}I emission due to biomass burning during this month can be roughly estimated from the CH$_3$I/CO molar emission ratio (1.3×10^{-5}) (Bell et al., 2002) combined with aboveground biomass density of 20.7 kg/m^2, CO emission factor (110 g/kg) and combustion factor (0.48) typical for tropical forest (Freitas et al., 2005). Around 0.02 Gg of CH$_3$I were emitted during this month, 0.7% of global emissions due to biomass burning. Other gaseous emissions could not be estimated and particulate emission was roughly estimated as 25% from gas to particulate ratio 3:1 for trees (Amiro et al., 1996). Assuming that the burned forest presents an isotopic ratio of around 5×10^{-10}, about 15 mg of ^{129}I was injected in the air during this month. The NOAA Hybrid Single-Particle Lagrangian Integrated allowed us to link the biomass burning source in Brazil with the receptor in Posadas (Draxler and Hess, 1998). We present a rough estimate of the transport, dispersion, and wet and dry deposition over Posadas (Fig. 5). We assume a constant ^{129}I emission rate (0.02 mg/h) during the entire month from a point source placed at (Lat.: 5°S; Lon: 45°W; Altitude: 1500 m a.s.l.). Precipitation in Posadas had to be adjusted by a factor 30 to match with meteorological data provided by Servicio Meteorológico Nacional Argentino.

Estimated daily deposition $d_D(t)$ of ^{129}I atoms/m^2 onto Posadas may be expressed as a daily contribution to the ^{129}I content in pasture $C_{pD}(t)$ (per kg) using the fractional of aerial deposition retained on plant $R = 0.25$ and the plant yield $Y = 1.8$ kg/m^2 by means of the following equation (Soldat, 1976; NCRP, 1983).

$$C_{pD}(t) = \frac{R \cdot d_D(t)}{Y} \tag{1}$$

The total content in pasture $C_p(t)$ is modulated by daily deposition C_{pD} and iodine ecological half-life on pasture ($T_{Eco} = 14$ d). Thus, $C_p(t)$ can be estimated as

$$\frac{dC_p}{dt} = \frac{dC_{pD}}{dt} - \lambda_{Eco} \cdot C_p(t). \tag{2}$$

Daily contribution to ^{129}I in thyroid $Q_D(t)$ can be known from the daily intake of fresh forage for a bovine ($M = 40$ kg) and the fraction $f = 0.3$ of this iodine reaching the gland.

$$Q_D(t) = f \cdot C_p(t) \cdot M \tag{3}$$

The total ^{129}I concentration in thyroid Q is a result of the daily intake and the loss, estimated using the biological half-life ($T_{Bio} = 100$ d).

$$\frac{dQ}{dt} = \frac{dQ_D}{dt} - \lambda_{Bio} \cdot Q(t) \tag{4}$$

Results are shown in Fig. 6 for total content in pasture $C_p(t)$ (a) and total ^{129}I concentration in thyroid $Q(t)$ (b), respectively.

This model indicates that 25% of the ^{129}I concentration measured in the sample from Posadas (6×10^{10} ^{129}I atoms in thyroid) comes from the biomass burned in Pará and Maranhão during November, supporting the initial hypothesis. Biomass burning from other months and other locations should be taken into account to obtain a complete estimation of the contribution of ^{129}I recycling due to biomass burning. The estimated deposition onto Posadas during November (2×10^7 ^{129}I atoms/m^2) can be compared with the estimation of annual meteoric flux of 15 g/year onto continental USA, which corresponds to 9×10^9 ^{129}I atoms/m^2/year (Moran et al., 1999).

Another ^{129}I recycling-source could be volatilization of iodine in Amazonas. Iodine that has been deposited onto the continent can be volatilized from plants and soils (Sive et al., 2007; Fuge, 1990). Similar behavior was reported after a rainwater isotope composition study

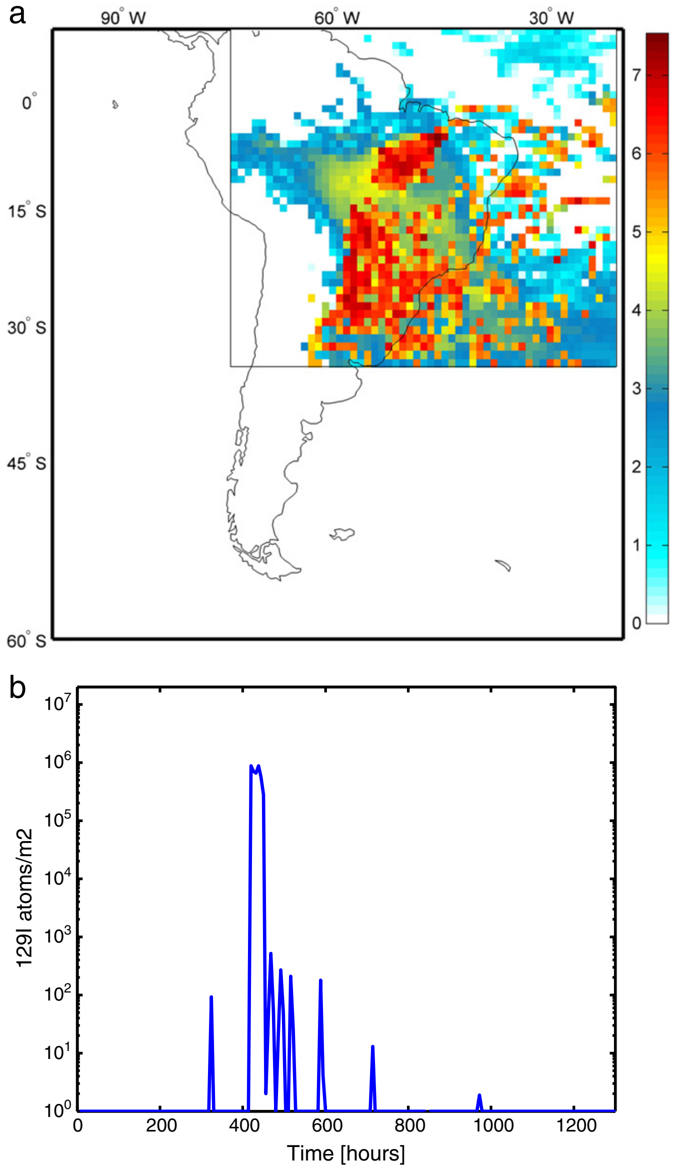

Fig. 5. (a) Integrated ^{129}I deposition (10^^{129}I atoms/m^2) from November 1st to December 15th of 15 mg uniformly emitted during November 2009 from a point-source using NOAA-HYSPLIT regional dispersion model. (b) Daily deposition $d_D(t)$ onto Posadas during the same period. Deposition values were corrected by a factor 30 to match with precipitation data provided by Servicio Meteorológico Nacional Argentino.

done in Northern Argentina, for ^{2}H and ^{18}O (González et al., 2009). It was well identified that during Summer there are precipitation events associated to recharged rain-water in the Amazonas.

The observed patterns of seasonality rule out high altitude interhemispheric ^{129}I transport which is most effective in mass exchange from Northern to southern hemisphere during the months of June and August (Newell et al., 1969).

3.1. Comparison with previous data

The only previous work reporting ^{129}I levels in thyroids from the Southern Hemisphere (Handl, 1996) estimated ^{129}I/^{127}I levels in the mid-region of Chile for the years 1985 through 1986 ($1.0-4.2\times10^{-9}$), and in South Eastern Australia for the years 1957 and 1989 ($5-6\times10^{-10}$). These values are up to one order of magnitude larger than our highest value despite the fact that the global surface inventory of ^{129}I was significantly lower at the time. All those measurements, however, were made via Neutron Activation Analysis (NAA) techniques, which are limited in sensitivity to 10^9 atoms/g of ^{129}I (Hou et al., 2009). The ^{129}I/^{127}I ratios measured in river samples from Santiago (Chile) and Iguazú Falls (Argentina/Brazil) (Snyder and Fehn, 2004) are in the range of our measurements.

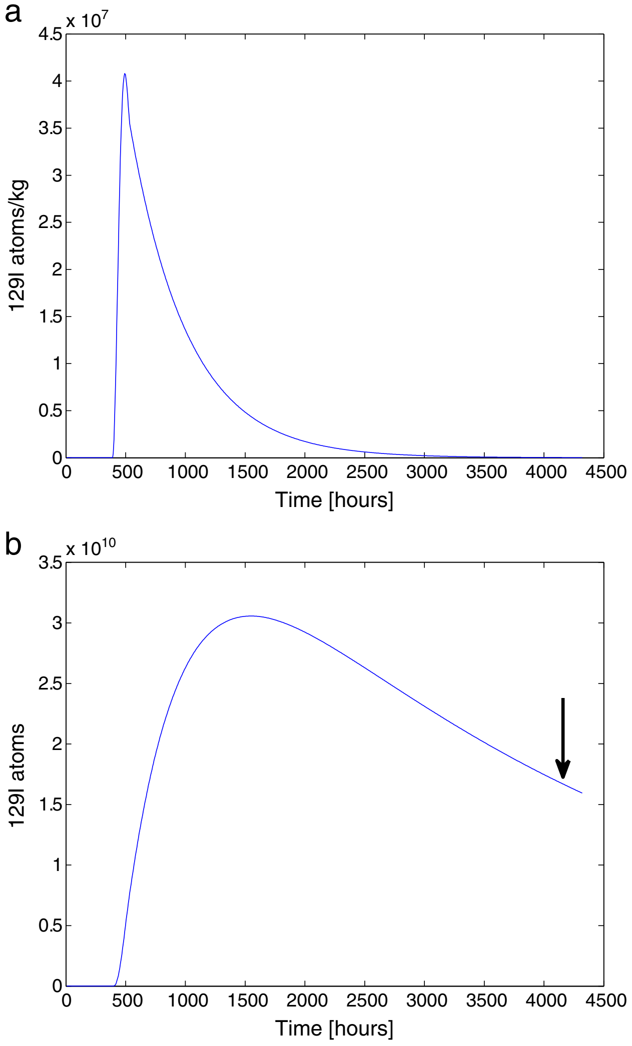

Fig. 6. Content of ^{129}I in pasture C_p (a) and concentration in thyroid Q (b) from November 1st 2009 to mid-April 2010 due to biomass burning in Pará and Maranhão during November 2009. The arrow indicates the date when the sample was taken.

In most of the cases, our ^{129}I/^{127}I ratios are lower than values measured in the Northern Hemisphere, including also those stemming from areas far away from the nuclear fuel reprocessing plants of Cape de La Hague (France) and Sellafield (UK), which are known as the major ^{129}I sources. For example, the average ^{129}I/^{127}I ratio in Taiwanese thyroids of grazing animals is about 1×10^{-9} (Chao and Tseng, 1996), while the mean ^{129}I/^{127}I ratio in Chinese human thyroids is 1.1×10^{-9} (Hou et al., 2000).

Samples from Southern Patagonia result in similar ^{129}I/^{127}I ratios as the ones from animal thyroid gland powder, sampled in 1943 in USA. This pre-anthropogenic sample, measured via AMS, yielded values of $(7.0\pm1.5)\times10^{-12}$ (Schmidt et al., 1998) and $(4.6\pm0.5)\times10^{-12}$ (Szidat et al., 2000). There are other pre-anthropogenic values reported, measured by means of less sensitive NAA, which are higher by at least one order of magnitude (see above). The ^{129}I levels found in Argentina, much lower than the values obtained from other regions or even close to pre-anthropogenic ^{129}I/^{127}I ratios, might imply the absence of local sources.

4. Conclusions

^{127}I and ^{129}I concentrations in animal thyroids from Argentina were determined by means of GC and AMS, respectively. This was the first determination of ^{129}I in this particular matrix in the southern hemisphere via the extremely sensitive AMS technique. The isotopic ratios measured in this work are generally lower than those found in thyroids from the Northern hemisphere, even for those Northern areas far from known ^{129}I-sources. In fact, two sampling sites present ^{129}I/^{127}I ratios similar to pre-anthropogenic values reported previously (Schmidt et al., 1998; Szidat et al., 2000). This might imply the absence of local sources in Argentina.

Our data suggest a strong dependence with latitude and season that can be understood by accounting for both probable sources and the deposition pathways of the two iodine isotopes.

The collected data can be used as reference values for Argentina and will contribute to the analysis of the global cycle of the ^{129}I which is scarcely known because of lack of data on ^{129}I concentration levels and the complex behavior of this radioisotope in the environment.

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