^{129}I Dispersion in Argentina: Concentrations in Fresh and Marine Water and Deposition Fluences in Patagonia

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ABSTRACT: Measurements of total iodine (I) and iodine-129 (^{129}I) concentrations in rivers and lakes of Argentina are presented. Their latitudinal distribution can be explained by taking into account their main sources (oceanic emissions and biomass burning for I, and atmospheric nuclear tests for ^{129}I), transport mechanisms, and fallout patterns. From the measured ^{129}I concentrations in the studied lakes, deposition fluences for their catchment areas were estimated. These results agree with a model of the global deposition pattern due to the ^{129}I released by atmospheric nuclear weapon tests and with other fluences reported for the southern hemisphere. In addition, the first measurements of ^{129}I in shallow seawater from the South Atlantic Ocean are presented and discussed.

Iodine and 129-Iodine in Argentinean Lakes
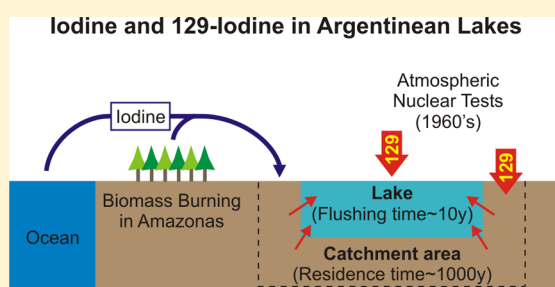
1. INTRODUCTION

129-iodine is a long-lived radionuclide ($T_{1/2}$ = 15.7 My) with a natural hydrospheric inventory of 140 kg (11 kg in the nonmarine hydrosphere) and a natural ^{129}I/I isotopic ratio close to 1.5×10^{-12}.[1,2] Since 1945, human activities increased its hydrospheric inventory significantly: nuclear weapon tests injected around 90 kg to the atmosphere, increasing the ^{129}I/I ratio in the shallow ocean to $\sim 5 \times 10^{-11}$.[1] More significant, nuclear fuel reprocessing plants placed in the northern hemisphere released to the atmosphere and the ocean more than 5400 kg for the period 1948−2010. In this way, the ^{129}I/I isotopic ratio was enhanced up to 6 orders of magnitude above the natural background in certain areas of the northern hemisphere.[3]

In the southern hemisphere, nowadays, natural sources and nuclear tests have a similar contribution in every ^{129}I surface compartment inventory (i.e., atmosphere, ocean mixed layer, surface soil region, and biomass).[4] Isotopic ratios of samples stemming from the southern hemisphere range between the natural value and the one related with nuclear tests.[1,5]

Argentina has received mainly stratospheric fallout from the nuclear tests events carried out in the northern hemisphere, whereas the nuclear tests in the southern hemisphere from 1966 to 1974 induced tropospheric fallout.[6] Recent nuclear activities in this hemisphere are constrained to one nuclear power plant in South Africa, two in Brazil, and two in Argentina; these last two countries have a third plant under construction each. There are no nuclear reprocessing plants for the austral hemisphere.

Recently, we published measurements of I and ^{129}I content in cattle thyroids stemming from different regions of Argentina.[5] The measured ^{129}I/I ratios, ranging from 3.1×10^{-12} to 3.8×10^{-10}, were smaller than those typical for the northern hemisphere. In fact, the samples from southern Patagonia present ^{129}I/I ratios compatibles to natural values. Moreover, we reported that I and ^{129}I contents presented a clear dependence with latitude, which was understood taking into account sources (oceanic emissions and biomass burning), transport mechanisms, and deposition pathways of the iodine nuclides.

These works were done as part of a recent project to add more experimental data of ^{129}I concentrations in the southern hemisphere. It is also aim of this work to contribute to the understanding of the ^{129}I sources and environmental distribution in Argentina, since there is scarce information concerning the presence of this radioisotope in the austral hemisphere. In the present article, the ^{129}I content in surface and marine water from Argentina was studied. Sampling and analytical procedures are described in Section 2. The obtained results are presented and discussed in Section 3.

2. EXPERIMENTAL METHOD

Water samples (between 2 to 6 L volume each) were collected in plastic bottles between December 2010 and April 2011 (for the sampling locations see Figure 1). Marine samples (1 L)
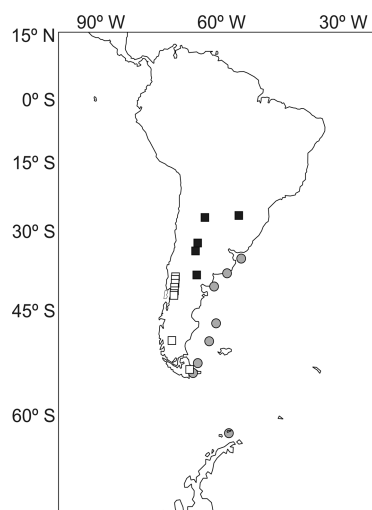

Figure 1. Map of sampling locations studied in this work. White squares correspond to lakes, black squares correspond to rivers, and gray circles correspond to shallow seawater.

from Argentinean Sea platform were obtained by the Puerto Deseado Oceanographic Ship during January 2011. The Antarctic marine sample stemmed from Bransfield Strait (1 L), between the South Shetland Islands and the Antarctic Peninsula. Lake samples were taken off-shore.

Particulate matter was removed using a 0.45 μm filter. The total iodine (I) content was determined using ICP-MS (inductively coupled plasma mass spectrometry) following the operating conditions of Tagami and Uchida.[7] In the case of seawater, we diluted the samples (1:10) for iodine determination. The detection limit for the ICP-MS was 0.02 μg/L.

The samples were chemically treated at the TANDAR Laboratory in Buenos Aires following the procedure described in ref 8. Inorganic iodine and an unknown low fraction of organic iodine were extracted from the samples using a chloroform method and were finally precipitated as AgI.

This fact has to be taken into account because organic iodine accounts up to 50% of the iodine in freshwater samples and up to 10% in seawater.[9] Therefore, measured ^{129}I concentrations and their corresponding ratios to total iodine reported for freshwater in this work may be underestimated by a factor up to 2 in the worst case scenario, as the reported values did not quantitatively include organo-iodine.

To quantify a potential ^{129}I contamination that the chemical treatment of the samples might has introduced, blank samples were prepared from the carrier material following the same procedure.

The water samples were reduced to a final volume of 200 mL under controlled pH values of ~9−10 using NaOH after adding 700 μg of Woodward iodine (700 μg/mL), which served as carrier. The iodine from Woodward is a commonly used blank material, since its low ^{129}I/^{127}I ratio (2×10^{-14}) is well determined.[10] This value was confirmed in the present work. All iodine species were first converted to iodide and then oxidized to I_2. Iodine was extracted in $CHCl_3$, and the aqueous phase was discarded. Iodide was re-extracted in water. This extraction, concentration, and back-extraction was repeated twice for purification. The iodide was combined with $AgNO_3$ to precipitate the AgI. The precipitate was centrifuged and washed with water three times. The resulting AgI was dried at 70 °C for 10 h, mixed with silver powder, and then pressed into a Cu sample holder.

The AMS measurements were performed at the Heavy Ion Accelerator Facility (HIAF) at the Australian National University, which is based on a 14UD pelletron accelerator.[11] We used a 32 sample MC-SNICS ion source and sputtered the sample with a Cs beam. Negative ions (I$^-$) were extracted and accelerated to 155 keV. The negative ions extracted from the source were deflected 90 degree from horizontal into vertical direction and injected into the tandem accelerator. For ^{129}I measurements, we ran the accelerator at a terminal voltage of ca. 11 MV. We selected the 7+ charge state with the double focusing 90 degree analyzing magnet, which bends the beam back into horizontal direction. After passing a switching magnet and a Wien filter, the rare radioisotope ^{129}I^{7+} was counted in a multianode ionization chamber. The beam intensity of stable ^{127}I was measured both at the low energy side and at the high energy side with Faraday cups. The ratio measurements were performed in sequential mode; that is, we measured ^{127}I

Table 1. Total Iodine and ^{129}I Concentrations in Water Samples As Well As the Resulting ^{129}I/I Ratios Measured in Rivers (R) and Lakes (L)

sampling site	latitude [deg]	longitude [deg]	date	total iodine [μg/L]	^{129}I [10^5 atoms/L]	^{129}I/I [10^{-11}]
Paraná (R)	−27.3	−55.9	12/2010	17.43 ± 0.44	5.2 ± 1.1	0.63 ± 0.15
Dulce (R)	−27.7	−64.2	3/2011	27.82 ± 0.64	7.5 ± 1.1	0.572 ± 0.084
Grande (R)	−33.0	−66.1	3/2011	7.15 ± 0.18	2.40 ± 0.34	0.71 ± 0.12
Atuel (R)	−34.5	−66.5	2/2011	3.49 ± 0.11	3.3 ± 1.2	1.99 ± 0.79
Negro (R)	−39.1	−66.2	3/2011	3.53 ± 0.17	2.72 ± 0.62	1.63 ± 0.45
Huechulaufquen (L)	−39.6	−71.3	3/2011	1.43 ± 0.12	1.04 ± 0.30	1.53 ± 0.57
Lácar (L)	−40.1	−71.5	3/2011	1.20 ± 0.12	2.21 ± 0.50	3.9 ± 1.3
Nahuel Huapi (L)	−40.8	−71.5	3/2011	2.78 ± 0.12	5.19 ± 0.86	3.94 ± 0.82
Mascardi (L)	−41.3	−71.5	3/2011	1.12 ± 0.13	4.04 ± 0.45	7.6 ± 1.7
Puelo (L)	−42.1	−71.6	3/2011	2.08 ± 0.20	1.44 ± 0.22	1.46 ± 0.36
Menéndez (L)	−42.6	−71.8	3/2011	1.41 ± 0.10	4.50 ± 0.48	6.7 ± 1.2
Futalaufquen (L)	−42.8	−71.7	3/2011	2.13 ± 0.11	5.04 ± 0.46	5.09 ± 0.73
Argentino (L)	−50.2	−72.3	4/2011	2.81 ± 0.11	6.31 ± 0.62	4.75 ± 0.64
Fagnano (L)	−54.5	−68.0	3/2011	10.43 ± 0.44	7.27 ± 0.72	1.47 ± 0.21

currents for 10 s, switched then to ^{129}I counting mode for 2 to 3 min, and then switched back to the ^{127}I current measurement. Such sequences were repeated twice. Switching between the different isotopes was done automatically. This AMS system allows measurements with an accuracy between 2 and 4%. All isotope ratios of the processed samples were at least 1 order of magnitude above the identically treated chemical blank samples, $(2.7 \pm 0.9) \times 10^{-14}$.

3. RESULTS AND DISCUSSION

3.1. Freshwater. Table 1 summarizes the experimental results for the freshwater samples. The sampling sites, ordered from North to South, correspond to five rivers and nine Patagonic lakes. Iodine content ranged from 1 to 28 μg/L. The ^{129}I concentrations obtained from the AMS measurements are within the 10^5-10^6 atoms/L. Uncertainties of blank, sample, and standard measurements were dominated mainly by counting statistics. The uncertainties given are 1σ. In what follows, an analysis of these results as well as a comparison with measurements in other regions will be done.

The content of total iodine in samples is comparable to values previously reported.[7,12] The latitudinal dependence is plotted in Figure 2. Values decrease southwards down to
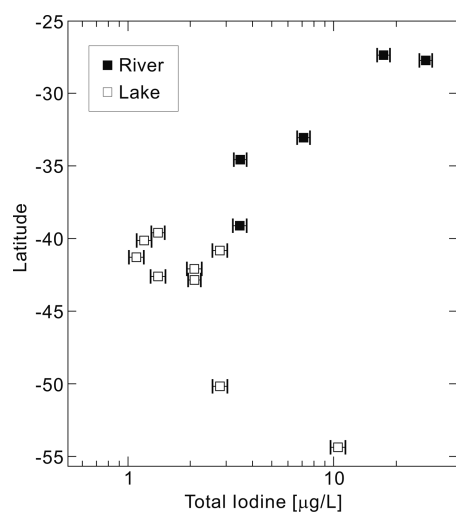

Figure 2. Latitudinal profile for total iodine content in the samples. It can be understood by considering the main iodine sources for this region and atmospheric transport and deposition pattern. See text for details.

around $-40°$. In sites more southern than $50°$, higher iodine concentrations were found again. This latitudinal dependency can be explained by considering the main sources of atmospheric iodine for South America[13] and the regional winds and precipitations patterns,[14] which eventually wash out the iodine down to the surface. Precipitation rates roughly decrease from northern to southern Argentina. At Northern and Central Argentina, the main path for atmospheric iodine are the Atlantic cyclonic winds which transport iodine stemming from emissions at the Equatorial Atlantic Ocean or from biomass burning in the Brazilian Amazonia. In the southern part of Argentina, emissions from the South Pacific and the South Atlantic are the most relevant sources. The intense western airstream allows the incoming of Pacific air over the southern Andes Mountains (the height of which is

much lower than in Central Argentina), with sporadic and local incursions of Atlantic air.[14]

The iodine latitudinal profile is quite similar to the one reported for iodine content in animal thyroids from Argentina.[5] In that work, the latitudinal profile could be explained assuming an atmospheric origin for the total iodine. The similar behavior in the profile between those thyroid samples and these water samples suggests that the main contribution of the iodine content for the studied watersheds in the present work might be also atmospheric.

On the other hand, the content of the radioisotope ^{129}I in the samples studied in this work are between 1.0×10^5 atoms/L and 7.5×10^5 atoms/L, which lie between the natural level (3.7×10^4 atoms/L) and the estimated level due to the nuclear tests (1.0×10^6 atoms/L). These concentrations are lower than others reported for the same region[1,10] and at least 1 order of magnitude lower than ^{129}I levels measured in samples from the Northern Hemisphere, even for samples collected far away from the nuclear fuel reprocessing plants.[15]

The latitudinal profile for ^{129}I is shown in Figure 3. It does not present a clear dependency with latitude as observed for total iodine, although lower values of this radioisotope are found at around $-40°$.
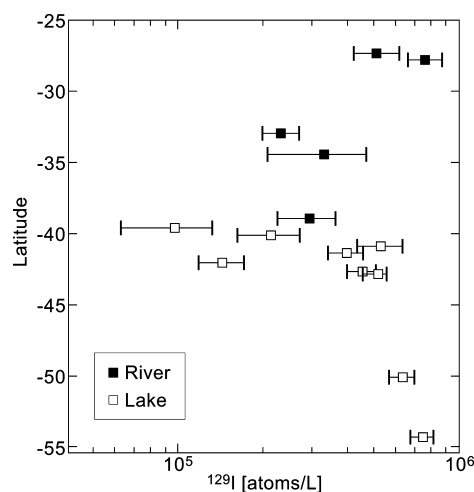

Figure 3. Latitudinal profile of the ^{129}I concentration for freshwater. Values lie between the natural (3.7×10^4 atoms/L) and nuclear weapon tests (1.0×10^6 atoms/L) levels.

For the southern region, these water samples yield $^{129}I/I$ ratios similar to the level associated with nuclear weapon tests (5.0×10^{-11}), while animal samples from the same region have shown $^{129}I/I$ ratios up to 1 order of magnitude lower, close to the natural ratio $(4 \pm 1) \times 10^{-12}$.[5] This marked difference will be discussed later (see paragraph four in Section 3.2).

Table 2 shows an estimation of the ^{129}I deposition fluences spatially averaged over the catchment area of the studied lakes obtained from the measured concentrations quoted in Table 1. To convert the concentrations into fluences, a model of lake water exchange presented in ref 16 was used. In that work, the authors assumed that rainwater deposited in the catchment area could be transport to the lake by means of direct deposition onto lake's surface and river inflow (which is a delayed signal). The same model was successfully applied in ref 10 for the Southern Hemisphere (a) considering an actual low ^{129}I flux for this hemisphere (neglecting current direct deposition contribution) and (b) assuming that the fraction of iodine

Table 2. Measured and Calculated Deposition Fluences for the Studied Lakes[a]

lake	lat [deg]	lon [deg]	t_F [y]	A_C [km^2]	A_L [km^2]	V_L [km^3]	P_R [mm/y]	measured deposition fluence (τ = 4000 y) [10^{11} atoms/m^2]	measured deposition fluence (τ = 1000 y) [10^{11} atoms/m^2]	calcd depostion fluence [10^{11} atoms/m^2]
Huechulaufquen	−39.6	−71.3	4.96	(963)	78.2	11.1	900	9.0	2.3	4.3
Lácar	−40.1	−71.5	5.8	1048	49.0	8.1	1380	11.6	2.9	8.2
Nahuel Huapi	−40.8	−71.5	12.3	4260	557	87.4	950	18.0	7.1	6.4
Mascardi	−41.3	−71.5	3.72	250	39.2	4.4	2100	74.1	18.5	11.4
Puelo	−42.1	−71.6	1.0	3040	44	4.9	1750	9.3	2.3	9.9
Menéndez	−42.6	−71.8	2.8	715	55.7	8.3	2300	74.3	18.6	12.3
Futalaufquen	−42.8	−71.7	0.94	2920	44.6	4.5	2300	33.1	8.3	12.3
Argentino	−50.2	−72.3	(10)	17400	1466	219.9	210	24.7	7.5	2.6
Fagnano	−54.5	−68.0	42	3537	590	46.8	600	1.6	1.2	4.0

[a]Measured deposition were calculated using the soil residence time of iodine τ reported in ref 17. (4000 y) and the best-fitting value τ = 1000 y estimated for ^{129}I in this work (see text for details). The table also contains limnological parameters (flushing time t_F, catchment Area A_C, lake surface A_L and lake volume V_L) and the annual precipitation rates (P_R) of the studied catchment areas. Values in brackets were estimated by us, all others were given from local governmental agencies (Sub-secretaría de Recursos Hídricos and Servicio Meteorológico Nacional, Argentina).

Table 3. I and ^{129}I Concentrations and ^{129}I/I Isotopic Ration in Marine Water as Measured in This Work[a]

latitude [deg]	longitude [deg]	date	iodine [μg/L]	^{129}I [10^5 atoms/L]	^{129}I/I [10^{-12}]	temp. [°C]	salinity [PSU]
−36.0	−55.3	01/2011	34.1 ± 3.2	40 ± 11	24.7 ± 9.1		
−38.8	−58.8	01/2011	30.2 ± 3.3	39 ± 13	27 ± 12		
−41.2	−62.0	01/2011	30.3 ± 2.1	42 ± 12	29 ± 10	19.79	34.004
−47.5	−61.5	01/2011	35.0 ± 2.4	4.9 ± 1.9	3.0 ± 1.3	12.84	33.281
−50.0	−63.2	01/2011	34.3 ± 2.4	4.6 ± 2.0	2.8 ± 1.4	12.12	33.132
−53.5	−66.0	01/2011	33.4 ± 3.8	9.0 ± 2.8	5.7 ± 2.4		
−54.9	−67.2	01/2011	33.1 ± 2.0	6.0 ± 2.1	3.8 ± 1.6	9.26	31.275
−62.3	−58.3	02/2011	34.8 ± 2.5	5.3 ± 1.3	3.2 ± 1.0		

[a]Also given are temperature and salinity of the collection sites (where available).

deposited by rain within the catchment area reaching the lake water body is really low. Therefore, this simple model presupposes that the ^{129}I inventory of the studied lakes depends only on the deposition fluence due to nuclear atmospheric tests: The ^{129}I fallout from those tests were adsorbed by the soil, and only a very small can be flushed out again during rain events and reach the lake water body. These ^{129}I can retardedly reach the lake water body after a mean residence time in the soil, τ. In those works and in the present study, a residence time originally reported for stable iodine (τ = 4000 y)[17] was used, although it is not clear if both iodine isotopes should have the same soil residence time (see text following). To convert ^{129}I measured concentrations into fluences, the model also needs some limnological parameters (flushing time t_F, catchment area A_C, lake surface A_L, lake volume V_L) of the studied areas. All those parameters are listed in Table 2. With the exception of the Fagnano Lake, the most southern lake sampled, the measured deposition fluences are in the order of 10^{12} atoms/m^2.

Table 2 presents the ^{129}I fluences calculated from the box model, developed in ref 16, that describes the global atmospheric transport and deposition of anthropogenic ^{129}I.[16] These calculated fluences were corrected for the local precipitation rate P_R, as is mentioned in that article. Although the model tends to underestimate the measured fluences, there is a fair agreement within 1 order of magnitude. Both the estimation of the ^{129}I deposition fluences from ^{129}I concentrations and the simulated global deposition pattern describe the behavior of complex systems that depend on several imprecisely known values (for example: lake description and atmospheric transport parameters, etc.). Besides, some aspects that may be relevant (characteristics of the studied soils and

environments, re-emission processes, etc.) were neglected in this analysis. Nevertheless, a significant improvement in the agreement between measured and calculated deposition fluences is achieved assuming a best-fitting residence time of ^{129}I τ = 1000 instead of the value τ = 4000 y reported by Kocher for iodine.[17] The ratio of the sum of the residuals of the values yield 0.22. Similar adjustment was done for the deposition fluences data set of ref 10. giving a value of τ = 750 y. These τ are about four times shorter than the reported for stable iodine (τ = 4000 y).[17] A shorter residence time indicates a higher mobility for this radionuclide. The authors from ref 18 reported experimental evidence that ^{129}I migrates faster than ^{127}I for all the studied soils, which presented different characteristics and hydrodynamical conditions. The difference in the residence time could be explained on the basis of their different probabilities of sorption due to different time periods for their input.[18]

Measured deposition fluences obtained in the Patagonian lakes are similar to other fluences reported for the Southern Hemisphere.[10,19] Furthermore, deposition fluences reported in this work agree reasonably well within 1 order of magnitude with the integrated deposition at the Fiescherhorn Glacier (Swiss Alps), which was (8.7 ± 0.5) × 10^{11} atoms/m^2 for the period 1950−1969.[20] Our values, however, are well below the (1.5 ± 0.1) × 10^{13} atoms/m^2 value estimated for the period between 1950 and 2002 for the same glacier.[20] All of our data clearly exceed the natural level of deposition fluences, (6.0 ± 1.5) × 10^{10} atoms/m^2 reported for a Russian soil sample from 1939.[21]

3.2. Seawater. The results of I and ^{129}I contents for samples originating from the shallow seawater (5 m depth) are listed in Table 3. Sampling stations are ordered from north to

south. Excluding the one from Antarctica, the water masses sampled correspond to a mix of subantarctic (Malvinas current) and subtropical (Brazil current) with continental discharges. Their relative ratios vary with latitude and season. One should point out that exchange from northern to southern hemisphere is not expected to be present at sea surface in this region. Water transport from the Northern Hemisphere is accomplished at deep level by the thermohaline circulation (North Atlantic Deep Water, NADW).

The iodine content is quite similar in all the studied samples, which is consistent with reports for iodine in the southern Atlantic Ocean.[22] Their concentrations are systematically lower (30−35 μg/L) than the oceanic mean iodine concentration (60 μg/L).

On the other hand, concentrations of the ^{129}I radioisotope present an interesting structure (see Figure 4). Samples in the
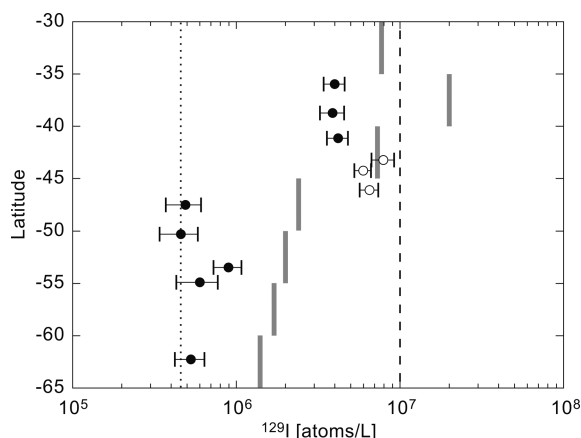

Figure 4. ^{129}I content in shallow seawater (black circles). Also, it is shown previously reported data for the Indian Ocean (white circles) [Povinec et al., 2011] and gray segments correspond to average for Δ = 5° latitudinal bands calculated by Snyder et al. (2010). The dotted line indicates the natural level (4.4 × 10^5 atoms/L), and the dashed line indicates the estimation for the level due to the nuclear weapon tests (1 × 10^7 atoms/L).

range from 36° to 41° have an almost constant concentration of ~4 × 10^6 atoms/L, while levels for samples stemming from 47° to 62° are almost 1 order of magnitude lower, ranging between 4 × 10^5 and 9 × 10^5 atoms/L. While the first set are slightly lower than the estimated when the contribution due to nuclear tests are considered (1 × 10^7 atoms/L), ^{129}I content in the southern samples were close to the natural level (4.4 × 10^5 atoms/L).[1] The mean isotopic ^{129}I/I ratios for the second set is (3.7 ± 1.5) × 10^{-12}, which is a above the natural ratio (1.5 × 10^{-12}).[2] In Figure 4, concentration values are compared to shallow samples from the Indian Ocean, which yield from 6.0 × 10^6 to 7.9 × 10^6 taken at 43−46°.[23,24]

In ref 25, it was pointed out that the iodine present in the edible part of a plant has an exclusive atmospheric origin. In ref 5, based on the iodine oceanic emission rates and wind patterns in southern Patagonia (sampling latitude: −51.5°), we suggest that the iodine isotopes found in cattle thyroids, after pasture intake, come predominantly from iodine emissions of the Pacific and Atlantic Oceans. In this sense, it is remarkable that the isotopic ratio average of those thyroids reported in ref 5, (4 ± 1) × 10^{-12}, is compatible with the seawater ratio (sampling latitudes: −53.5° and −54.9°) (4.8 ± 2.0) × 10^{-12} reported in this work.

One way to visualize the magnitude of the ^{129}I concentration values obtained in this work is to compare them with average ^{129}I concentrations for shallow seawater reported by ref 1. In that work, the authors calculated ^{129}I concentrations across 5° bands of latitude. Such a comparison is shown in Figure 4. The values reported in this work are slightly lower than the ones reported in ref 1. It should be noted, however, that their averages were derived by combining fresh and marine water.

In summary, in this article we present ^{127}I and ^{129}I concentrations measured in surface and seawater samples stemming from Argentina. Latitudinal dependence for ^{127}I in freshwater can be understood taking into account the main iodine sources for the region and the atmospheric transport and deposition pattern. ^{129}I contents in surface water are well above the natural level and close to values associated with atmospheric nuclear tests. ^{129}I deposition fluences in the studied lakes' catchment areas are similar to calculated ones using a simple atmospheric box model considering atmospheric nuclear explosions. This indicates that the fallout of these explosions is still the main source of ^{129}I (memory effects of atmospheric tests). Also, we present the first set of ^{129}I data from south Atlantic marine samples, with concentrations values lies between natural and nuclear tests levels.

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Notes

The authors declare no competing financial interest.

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