

Iodine

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1 SUMMARY

Iodine is an important element in studies of human nutrition to combat iodine-deficiency disorders, environment, and human-health protection from the anthropogenic release of radioactive iodine, and global-scale hydrologic processes. This article summarizes its basic chemistry, occurrence, and speciation in various environmental compartments, extraction, and analytical techniques, as well as the fate and transport behavior of different iodine species in the subsurface. Biogeochemical cycling of iodine in the environment is complex because iodine occurs in multiple oxidation states, and inorganic and organic species may be hydrophilic, atmophilic, and biophilic. Organically bound iodine can be a significant fraction of total iodine in the atmosphere, biosphere, hydrosphere, and lithosphere. In natural soils, nearly 90% of total iodine is present as organic species, while inorganic iodine becomes important (up to 50%) only in sediments with low organic matter content. Careful attention must be given to

iodine speciation and potential interconversion among species when interpreting the biogeochemical behavior of iodine in the environment. In addition to speciation, input concentration and residence time effects will influence the biogeochemical cycling of anthropogenic ¹²⁹I deposited on surface soils.

2 OCCURRENCE

Naturally occurring stable iodine is a single isotope with 74 neutrons and has mass number 127. Iodine was discovered in 1811 by French chemist Bernard Courtois when he sublimed the element from seaweed ash with concentrated sulphuric acid.^{1,2} The origin of the name comes from the Greek word *iodes* meaning *violet*. Iodine is a shiny, black, nonmetallic solid with a characteristic odor. Iodine sublimates easily as a gas, which is violet and intensely irritates the eyes, nose, and throat.

Iodine is a widespread trace element distributed in the atmosphere, lithosphere, hydrosphere, and biosphere at varying concentrations (Table 1) (*see Natural Radioactivity*). Iodine is an essential micronutrient in mammals (humans and animals) and is necessary for the production of thyroid hormones and for the proper functioning of the thyroid gland. Iodine deficiencies can lead to severe metabolic disorders, including thyroid enlargement (goiter), hypothyroidism (underactive thyroid gland), hyperthyroidism (overactive thyroid gland), mental retardation in infants and children whose mothers are iodine-deficient during pregnancy, reproductive damage, and childhood mortality.³ As reported by the World Health Organization,⁴ iodine deficiency is the world's most prevalent, yet easily preventable, cause of brain damage. Significant progress has been made since the primary intervention strategy for controlling iodine-deficiency disorders—universal salt iodization—was adopted in 1993. The number of countries where iodine deficiency is a public health problem has been halved over the past decade. However, 54 countries (approximately 40% of the world's population) are still at risk of iodine deficiency.⁴ Vitti *et al.*

also reported that 2.2 billion people worldwide are at risk of the disorders.³

In addition to concern about iodine-deficiency disorders, an uncontrolled release of radioactive iodine could constitute a direct threat to human populations because radioactive iodine is concentrated in the human thyroid.⁷ Iodine has one stable isotope, ¹²⁷I, with 42 recognized radioactive isotopes and isomers at atomic numbers 108–141 that include 10 fission products with very short half-lives ranging from minutes to a few hours (Table 2).^{2,8} The two radionuclides ¹³¹I and ¹²⁹I are of particular environmental concern. Following a nuclear accident (such as the one at Chernobyl) or an intentional radiological release, ¹³¹I would be an acute contaminant because of its short half-life of 8.02 days and a high specific activity of 4.59×10^3 TBq g⁻¹ (where TBq is terabecquerel). According to UNSCEAR, the ¹³¹I inventory in the reactor core at Chernobyl was estimated to be 3.2×10^6 TBq and the total ¹³¹I released during the course of the accident was about 1.7×10^6 TBq.⁹ Having a long half-life (1.57×10^7 years), ¹²⁹I is an important fission product with a fission yield of

Table 1 Compilation of total iodine and speciation in different systems^{(a)5}

System		Total concentration	Speciation
Atmosphere		1–100 ng m ⁻³	Particulate iodine; inorganic gaseous iodine (I ₂ , HI, HOI); organic gaseous iodine (CHI ₃ , CH ₂ I ₂ , CH ₃ CH ₂ CH ₂ I)
Lithosphere	Earth's crust	0.45 mg kg ⁻¹	Iodide, iodate, and organic iodine (mostly)
	Sedimentary rocks	1–2000 mg kg ⁻¹	
	Metamorphic and magmatic rocks	<0.1 mg kg ⁻¹	
	Metamorphic rocks ¹	0.06–4.03 mg kg ⁻¹	
	Rock-forming minerals ¹	<0.02–1.20 mg kg ⁻¹	
	Soils	0.5–40 mg kg ⁻¹ (1–3 mg kg ⁻¹ commonly)	
Hydrosphere	Ocean water	45–60 µg l ⁻¹	<1–25 µg l ⁻¹ iodide; 25–60 µg l ⁻¹ iodate
	Fresh water	1–3 µg l ⁻¹	
	Nonmarine surface water ¹	0.01–73 µg l ⁻¹ (average 5 µg l ⁻¹)	Iodide, iodate, and organic iodine
	Precipitation	1–6 µg l ⁻¹	
	Rain and snow ¹	0.5–20 µg l ⁻¹	
	Spring and well water ¹	3.5–1270 µg l ⁻¹	
	Subsurface brines and formation waters ¹	4–380 µg l ⁻¹	
Biosphere	Seaweeds	10–6000 mg kg ⁻¹	Mostly iodide; <5% iodate; 5–40% organic iodine
	Brown algae	100–6000 mg kg ⁻¹	
	Green algae ¹	21–130 mg kg ⁻¹	
	Fish and molluscs ¹	0.2–6.0 mg kg ⁻¹	
	Terrestrial plants	<1 mg kg ^{-1,5} 0.22–6.9 mg kg ⁻¹¹	
	Thyroid	50–5000 mg kg ⁻¹	Iodide (mainly); organic iodine
	Tissues other than thyroid	<1 mg kg ⁻¹	
	Breast milk in European women	95 ± 60 µg l ⁻¹	
	Urine ⁶	Optimal at 100–200 µg l ⁻¹ for iodine nutrition	

^(a) All iodine results for biological samples are based on mass per dry matter.

Table 2 Nuclear properties of iodine isotopes with half-life more than 10 min^(a)

Isotope	Half-life	Decay mode	E_{\max} (keV)
¹²³ I	13.27 h	EC + β^+	1074.9 (97%, EC)
¹²⁴ I	4.18 days	EC + β^+	2557 (25%, EC), 3160 (24%, EC), 1535 (12%, β^+), 2138 (11%, β^+)
¹²⁵ I ^(b)	59.41 days	EC	150.6 (100%)
¹²⁶ I	13.11 days	EC + β^+ , β^-	869.4 (32%, β^-), 1489 (29%, EC), 2155 (23%, EC)
¹²⁷ I	Stable	β^- , EC + β^+	
¹²⁸ I	24.99 months	β^- , EC + β^+	2119 (80%, β^-)
¹²⁹ I	1.57×10^7 years	β^-	144.4 (100%)
¹³⁰ I	12.36 h	β^-	587 (47%), 1005 (48%)
¹³¹ I	8.02 days	β^-	606 (90%)
¹³² I	2.30 h	β^-	738 (13%), 1182 (19%), 2136 (19%)
^{132m} I	1.39 h	IT, β^-	1483 (8.6%, β^-)
¹³³ I	20.8 h	β^-	1240 (83%)
¹³⁴ I	52.5 m	β^-	1307 (30%)
¹³⁵ I	6.57 h	β^-	970 (22%), 1388 (24%)

EC, electron capture; β^+ , positron emission; β^- , β emission; IT, internal transfer.

^(a)From Hou *et al.*¹⁰

^(b)¹²⁵I is produced by the electron capture decay of ¹²⁵Xe and is used in biological assays and in radiation therapy, as well as a biogeochemical tracer and yield tracer in laboratory testing and measurements of iodine.

0.9% from ²³⁵U (*see Uranium*) and 1.6% from ²³⁹Pu (*see Plutonium*).

With its long half-life, high-abundance fission yield, and presumably high mobility in the environment, ¹²⁹I has been recognized as one of the most important radionuclides in studies of environmental protection and human health, global-scale hydrologic processes, and nuclear nonproliferation. As summarized by Hu *et al.*, both natural and anthropogenic sources contribute ¹²⁹I to the environment (Table 3) (*see Anthropogenic Radioactivity*).¹¹ As the only naturally occurring radioactive isotope of iodine, ¹²⁹I is produced by cosmic-ray interactions with xenon in the upper atmosphere and by spontaneous fission of ²³⁸U in the geosphere (*see Natural Radioactivity*). Moran *et al.* reported that the total natural ¹²⁹I in the surface environment was about 80 kg, with just 5×10^{-4} kg in the atmosphere.¹²

Table 3 Major source terms of ¹²⁹I in the environment

Source	¹²⁹ I	
	(kg)	(TBq)
Natural hydrosphere ¹²	80	0.52
Natural atmosphere ¹²	0.0005	0.000003
Atmospheric testing ¹³	50–150	0.32–0.96
Chernobyl ¹³	6	0.04
Savannah River Site ¹¹	32	0.21
Hanford reservation ¹¹	260	1.7
NTS underground nuclear testing ¹¹	10	0.065
Spent fuel reprocessing (Europe), ^{10(a)}	5640	37
Potential Yucca Mountain repository ¹¹	14 400	94

^(a)Hou *et al.* reported a marine discharge of 5200 kg and atmospheric release of 440 kg, both by 2007.¹⁰

Nuclear-fuel reprocessing facilities constitute the major source (>90%) of ¹²⁹I released to the environment (Table 3). From 1966 to 1999, a total of 2300 kg (15 TBq) of liquid ¹²⁹I was discharged into the marine environment and another 64.2 kg (0.42 TBq) of gaseous ¹²⁹I was released into the atmosphere by La Hague in France.¹⁴ It is also reported that 2600 kg (17 TBq) of ¹²⁹I was discharged into the sea until 1998 by La Hague in France and Sellafield in England, an amount that is 50 times the total release from nuclear weapons tests and 3 orders of magnitude higher than the Chernobyl accident release.^{15,16} After 1998, ¹²⁹I has been discharged at a rate of about 0.2 and 1.2 TBq per year from Sellafield and La Hague, respectively.¹⁷ From these two European reprocessing facilities, the total release of ¹²⁹I was estimated to be about 255 and 4500 kg from gaseous and liquid discharges, respectively.^{13,18} In addition, two other facilities located in central Europe, such as Karlsruhe in Germany (operated during 1970–1989) and Marcoule in France (operated during 1959–1997), have released smaller amounts of ¹²⁹I into the environment of Western Europe. Gaseous emissions from the Marcoule facility were about 68 kg,¹³ whereas emissions from the Karlsruhe facility are considered negligible.¹⁹

From 1944 through 1972, the plutonium production operation at the Hanford Site in Washington released about 260 kg (1.7 TBq) of ¹²⁹I into the air.²⁰ By comparison, the operation of production reactors from 1953 to about 1990 at the Savannah River Site (SRS) in South Carolina released about 32 kg (0.21 TBq) of ¹²⁹I into the air, while the nuclear accident at Chernobyl released about 1–2 kg ¹²⁹I.^{12,15} The proposed geological repository for storing high-level nuclear waste at Yucca Mountain of Nevada could potentially contain as much as 14 400 kg (94 TBq) ¹²⁹I based on the storage of 70 000 tons of nuclear waste.¹¹

Anthropogenic inputs of ^{129}I have overwhelmed the natural inventory, increasing the hydrospheric $^{129}\text{I}/^{127}\text{I}$ ratio from 1.5×10^{-12} during the prenuclear era to up to 10^{-11} – 10^{-4} .^{14,21,22} This marked change in the ratio creates an opportunity for the application of anthropogenic ^{129}I as an environmental tracer, originally proposed by Edwards,²³ to examine a variety of processes including ocean circulation, biogeochemical cycling, and regional hydrologic processes.^{5,12,13,24,25}

The ^{129}I concentration in environmental samples (e.g., sea water, fresh water, rain, air, soils, sediments, plants, blood, milk, urine, etc.) has increased from 3 to 8 orders of magnitude compared with the prenuclear era level and has reached 10^{-10} – 10^{-4} for the $^{129}\text{I}/^{127}\text{I}$ ratio.¹⁰ Overall, the different levels of $^{129}\text{I}/^{127}\text{I}$ ratio in the environment are envisaged as 10^{-12} for the prenuclear era, 10^{-9} in slightly contaminated regions, and 10^{-9} – 10^{-6} in regions affected by releases from reprocessing plants. The highest ratios of $^{129}\text{I}/^{127}\text{I}$ (10^{-6} – 10^{-3}) have been found in the vicinity (<50 km) of reprocessing plants.¹⁰

The concentrations of ^{129}I in environmental samples are normally from 4 to 12 orders of magnitude lower than that of stable iodine.¹⁰ In addition, anthropogenic ^{129}I has only been participating in iodine cycling over the past 50 years, compared to the longer interaction times (thousands of years), as well as much higher concentrations of ^{127}I . Compared to ^{129}I , ^{127}I preferentially occupies the thermodynamically favorable sorption sites. Therefore, trace ^{129}I likely possesses a higher mobility than stable ^{127}I .^{26–28} Nevertheless, studies of both stable ^{127}I and radioactive ^{129}I serve complementary research purposes. Work on ^{129}I has generally been focused on its distribution and measurement, and species evaluation to some extent, in various environmental samples. Analysis of ^{129}I requires sophisticated analytical instrumentation that is not readily accessible to many researchers, and separation and purification from different matrices is labor intensive. On the other hand, studies of stable iodine can reveal insights regarding various biogeochemical processes of iodine cycling in the environment. This article provides a balanced summary of research on both ^{127}I and ^{129}I .

3 CHEMISTRY

The basic chemistry of iodine is presented in Table 4. While iodine is only slightly soluble in water, it dissolves in chloroform, carbon tetrachloride, or carbon disulfide to form beautiful violet-colored solutions.² Iodine will form compounds with most elements, but is less reactive than the other halogens, which displace it from iodide compounds and exhibits some metallic-like properties. Iodine compounds are important in organic chemistry and very useful in medicine (Figure 1). Iodine is biophilic and is particularly strongly concentrated in seaweeds from which it may be recovered. In

Table 4 Chemical properties of Iodine. (Reproduced from Ref. 2. © Taylor & Francis Group, 2000)

Name	Properties
Atomic number	53
Mass number of naturally occurring isotopes	127 (100%)
Atomic mass	126.9045
Electronic configuration	[Kr]4d ¹⁰ 5s ² 5p ⁵
Electronegativity	2.5
Color	Bluish-black for solid and liquid; violet for gas
Melting point	113.7 °C
Boiling point	184.4 °C
Critical temperature	546 °C
Atomic radius	133 pm
van der Waals's radius	215 pm
Molar volume	25.74 cm ³ mol ⁻¹ (at 20 °C)
Ionization energy	1008.7 kJ mol ⁻¹
Density	4.93 kg m ⁻³ (at 20 °C)

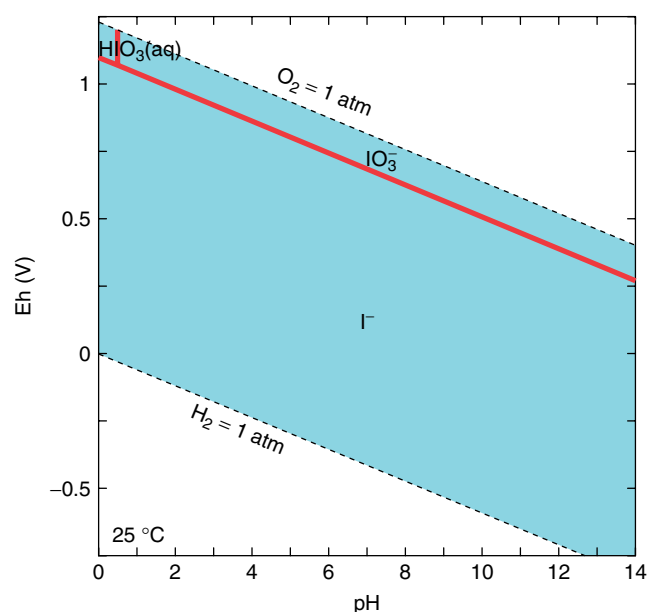


Figure 1 Eh–pH diagram, drawn at 25 °C and activity of 10^{-11} M, for iodine in a typical Na–K– HCO_3 type dilute water. Diagram produced using the “thermo.com.V8.R6+” database in the Geochemist’s Workbench (version 6.0)

inorganic forms, it is found at relatively high concentrations in Chilean saltpeter and nitrate-bearing caliche and in brackish waters and brines from oil and gas wells.²

Given its atmophilic and biophilic tendencies and large ionic radius that inhibits it from forming crystal structures, the biogeochemical behavior of iodine must be well understood before $^{129}\text{I}/^{127}\text{I}$ ratios can be used as an environmental tracer or geochronometer. With oxidation states ranging from -1 to $+7$ (Table 5) and a disparate set of associated species, iodine frequently exhibits complex

Table 5 Oxidation states in various substances

Oxidation state	Example substances
-1	I ⁻ (iodide), KI
0	I ₂ , I ₃ ⁻ , I ₅ ⁻
1	IO ⁻ (hypoiodite), ICl ₂ ⁻
3	IO ₂ ⁻ (iodite), ICl ₃
5	I ₂ O ₅ , HIO ₃ , IO ₃ ⁻ (iodate), IF ₅ , IF ₆ ⁻
7	H ₅ IO ₆ , H ₄ IO ₆ ⁻ , HIO ₄ , IO ₄ ⁻ (periodate), IF ₇

biogeochemical behavior and occurs as various species with their associated hydrophilic, atmophilic, and biophilic characteristics.¹ The predominant states of inorganic iodine in natural aqueous systems are -1 (iodide, I⁻) and +5 (iodate, IO₃⁻) (Figure 1). When Eh values are high (>1.2 V), some high-valence inorganic iodine, such as IO₄⁻ and HIO₄⁻, can be present.^{1,10} In reducing environments, aqueous iodine usually occurs as the mobile monovalent anion, I⁻. Under more oxidizing conditions, iodine is present as the more reactive IO₃⁻, which can lead to retarded transport through interaction with clays and organic matter;²⁹ sorption of iodine species on soils is discussed in Section 7.

4 SEPARATION TECHNIQUES

The choice of separation technique depends upon the sample matrix, the analysis method, the expected concentration of ¹²⁷I and/or ¹²⁹I, and the expected speciation.

4.1 Ion-Exchange Resin

The ion-exchange method was first published by Wilkins and Stewart.³⁰ After the addition of iodide carrier to the waters, all the iodine species are converted to iodide by sequential addition of NaOCl (sodium hypochlorite), NH₂OH·HCl (hydroxylamine hydrochloride), and NaHSO₃ (sodium bisulfite). The iodide is concentrated using anion-exchange chromatography resin (Bio-Rad AG1-X8) and eluted as iodate, using NaOCl solution. The eluate is treated with 0.1 M HNO₃, followed by 0.5 M NH₂OH·HCl, and the iodine is purified by extraction into chloroform and then back extraction into a dilute solution of Na₂S₂O₅ (sodium metabisulfite, which forms sodium bisulfate in solution). The extraction is repeated and the iodine is recovered by precipitation as silver iodide, which is then mixed with subequal quantities of niobium or silver powder for accelerator mass spectrometry (AMS) measurements. For non-AMS measurement of liquid samples, analysis may directly follow ion-exchange chromatography. Slightly modified methods of exchange resin for separation and purification have been published.^{31–34}

4.2 Alkali Fusion/Ashing

Sodium hydroxide is added to 1–10 g samples to reduce the temperature of fusion, followed by the addition of sodium peroxide to break organic bonds. Sample crucibles are heated in a muffle furnace for 30 min at 300 °C and for 2 h at 600 °C. Distilled water is used to leach the soluble fraction of iodine from the fluxed sample, and a mixture of 2N·H₂SO₄ and 1 M NaHSO₃ is used to extract insoluble periodate (IO₄⁻) present.³⁵ This method has the advantage of allowing relatively large sample sizes and produces a very low blank for preanthropogenic ¹²⁹I applications. Recovery, however, is variable and usually less than 80% and some forms of iodine may also be volatilized and lost during heating.

4.3 Pyrohydrolysis Combustion

A subgram solid-phase sample is placed in a quartz tube and heated to 1000 °C under oxygen flow as a pyrohydrolysis combustion. Evaporated iodine is collected in a trap solution containing tetramethyl ammonium hydroxide (TMAH) and Na₂SO₃. Concentrations of iodine are then determined by inductively coupled plasma-mass spectrometry (ICP-MS) or another suitable method for aqueous matrices.^{34,36} This method is advantageous because it can be applied to almost any solid matrix, including rock material with very low iodine concentration.³⁷ However, Gilfedder *et al.* have modified this process by trapping combusted iodine into Milli-Q water, and quantifying the extracts by As(III)–Ce(IV) UV/V photometry, wherein iodide catalyzes the oxidation of As(III) to As(V) and reduction of Ce(IV) to Ce(III).³⁸

4.4 Aqueous Extractions for Iodine Speciation Studies

Chemical extractions using KCl and/or TMAH (either sequential or single-step) were conducted to investigate iodine speciation (inorganic and organic) in soil and sediment samples.³⁹ Extraction of inorganic iodine from soils was performed by adding 10 ml of 5 mM KCl to approximately 2 g of soil sample. The mixture was shaken for 1 h on a shaker table and centrifuged at 5000 rpm for 30 min to remove particle sizes larger than 0.45 µm. The supernatant was decanted for analysis and an extraction with 10 ml of 5 mM KCl was repeated; both supernatants were analyzed by ion chromatography (IC) for iodide and/or iodate. This extraction scheme was selected based on an evaluation of the extraction efficiency determined by spiking iodine species into NIST SRM (National Institute of Standards and Technology Standard Reference Material) 2709 soil (San Joaquin), using three different extractants (deionized water, 0.05 M KCl, and 0.5 M KCl) and different iodine species.

Extraction of iodine contained in the organic components of the soils was performed by shaking 2 g of soil in 20 ml of 5% TMAH for 4 h, centrifuging, and determining

iodine by difference using IC and ICP-MS. Using the NIST SRM soils with well-known total iodine contents, extraction variables such as the temperature (TMAH extraction under either room temperature or 80 °C) on quantitative iodine extraction were also evaluated.³⁹

5 ANALYTICAL CHARACTERIZATION TECHNIQUES

Recently, Hou *et al.* summarized the various measurement techniques for ¹²⁹I in environmental samples, which are γ - and X-ray spectrometry, liquid scintillation counting (LSC), neutron activation analysis (NAA), AMS, and ICP-MS.¹⁰ This discussion regarding ¹²⁹I measurement is mainly from the work of Hou *et al.*,¹⁰ along with Table 6 that compares different analytical methods for ¹²⁹I. In addition, the analytical methods for stable iodine are described, as an understanding of the behavior of stable iodine is important for biogeochemical processes relevant for radioactive iodine.

5.1 γ - and X-Ray Spectrometry

Decay of ¹²⁹I emits β -particles with a maximum energy of 154.4 keV, as well as γ -ray of 39.6 keV and X-rays of 29–30 keV. γ - and X-ray spectrometry have been used to measure ¹²⁹I in thyroid, urine, seaweed, and nuclear waste samples using high-purity germanium or plenary silicon detectors.¹⁰ However, because of the low counting efficiency of γ -detection (<2%) and low γ -ray abundance (7.5%), as well as high background, the detection limit for γ - and X-ray spectrometry is as high as 20–200 mBq (Table 6).

5.2 Liquid Scintillation Counting

Decay of ¹²⁹I has a moderate β -energy, and a counting efficiency of 60–95% can be achieved with LSC. Using LSC and measuring ¹²⁹I separated from the matrix and other radionuclides result in a detection limit of about 10 mBq.¹⁰

Overall, there are only a few publications that report using LSC to measure ¹²⁹I.

5.3 Neutron Activation Analysis

Analysis of ¹²⁹I by NAA was first utilized in 1962 by measuring the decay of activation product ¹³⁰I (with a half-life of 12.3 h), which emits β -particles and γ -rays and has a higher specific radioactivity and suitable γ -energies.¹⁰ Hou *et al.* presented a diagram of the analytical procedure for the determination of ¹²⁹I by radiochemical NAA. Some nuclides (²⁴Na, ⁸²Br, ¹²⁸Te, ¹³³Cs, and ²³⁵U) will interfere with ¹³⁰I analyses, and purification steps are required. In addition, stable iodine can be determined by fast neutron reaction ¹²⁷I (n, 2n) ¹²⁶I.¹⁰ As reported by Gilfedder *et al.*, the detection limit for ¹²⁷I by commercially available NAA is not less than 1 mg kg⁻¹.³⁸

5.4 Inductively Coupled Plasma-Mass Spectrometry

Over the past two decades, ICP-MS has been widely used for both ¹²⁷I (mostly) and ¹²⁹I (rarely) because of increased accessibility of the instrumentation. However, low sensitivity, isobaric and molecular interferences (¹²⁹Xe particularly, ¹²⁷IH₂, ⁸⁹Y⁴⁰Ar, ¹¹⁵In¹⁴N, and ¹¹³Cd¹⁶O for ¹²⁹I), and memory effects (due to transformation between different species) have posed challenges for iodine analyses by ICP-MS. For example, iodine has a high ionization potential (10.45 eV), and its ionization efficiency in argon plasma is only about 30%. Several approaches have been proposed to mitigate the mass interferences and memory effect. A dynamic reaction cell that uses oxygen as the reaction gas has been found to significantly reduce the signal of isobaric xenon ions.^{40,41} Brown *et al.* also suggested that using 1% of tertiary amine solution as the sample matrix minimizes memory effect.⁴²

Although the detection limit for ¹²⁹I/¹²⁷I from direct water measurements using quadrupole ICP-MS is 10⁻⁶–10⁻⁵ (Table 6), which limits its application to samples with relatively high ¹²⁹I, ICP-MS has been widely used for the measurement of stable iodine.^{29,37,43–46} Furthermore, ICP-MS techniques have been employed to investigate iodine

Table 6 Comparison of measurement methods for the determination of ¹²⁹I. (Reproduced from Ref. 10. © Elsevier, 2009)

Detection method	Sample preparation	Detection limit	
		¹²⁹ I	¹²⁹ I/ ¹²⁷ I ratio
X- γ spectrometry	Direct measurement	100–200 mBq	10 ⁻⁵ –10 ⁻⁴
X- γ spectrometry	Separated iodine (AgI)	20 mBq	10 ⁻⁶ –10 ⁻⁵
LSC	Separated iodine	10 mBq	10 ⁻⁶ –10 ⁻⁵
RNAA	Separated MgI ₂ /I ₂ on charcoal	0.001 mBq	10 ⁻¹⁰
AMS	AgI	1 × 10 ⁻⁶ mBq	10 ⁻¹⁴
ICP-MS	Direct water measurement	0.04–0.1 mBq ml ⁻¹	10 ⁻⁶ –10 ⁻⁵
ICP-MS	Gaseous iodine	0.0025 mBq g ⁻¹	10 ⁻⁷

speciation after sequential extraction.^{39,47–49} Coupled with front-end chromatographic separation, ICP-MS determination of iodine provides direct measurement of iodine species, which is a powerful tool in examining the biogeochemical cycling of iodine in the environment.^{6,50–60}

5.5 Accelerator Mass Spectrometry

Only NAA and AMS are sensitive enough to analyze $^{129}\text{I}/^{127}\text{I}$ ratios in environmental samples (with ratios of 10^{-10} – 10^{-6}), and AMS is the only method for pre-nuclear age samples with $^{129}\text{I}/^{127}\text{I}$ ratios lower than 10^{-10} .¹⁰ The principles and application of AMS in ^{129}I analyses are summarized by Hou *et al.*¹⁰ The main limitation of AMS is the sample size requirement, which is between 0.1 and 1 mg I, even for systems with efficient ion sources and relatively high transmission efficiencies. Secondly, there are very few AMS facilities capable of running ^{129}I and these are not widely available for routine applications.

The first application of AMS for the analysis of ^{129}I in natural materials occurred in the spring of 1977 in the Nuclear Structure Research Laboratory (NSRL) at the University of Rochester in upstate New York. This facility, funded by United States National Science Foundation (NSF), commenced operations in 1966 and was decommissioned in 1995. The tandem accelerator mass spectrometer at NSRL produced the earliest ^{129}I data on environmental samples, directly counting atoms and increasing the sensitivity over counting methods by several orders of magnitude.^{61–65}

Available AMS facilities around the world that have a demonstrated capability for the analysis of ^{129}I include the following: the IsoTrace Tandetron AMS facility (closed in 2009) at the University of Toronto, Canada,^{66–69} the Purdue Rare Isotope Measurement Laboratory (PRIME Lab) at Purdue University in Indiana,^{5,12,21,25,70–73} 10-MV Center for Accelerator Mass Spectrometry (CAMS) at Lawrence Livermore National Laboratory in California,^{74–76} the 6-MV tandem accelerator at the PSI/ETH-Honggerberg in Zurich,^{77–80} 3-MV NSF-Arizona AMS Facility,^{81,82} Australian National University in Canberra,^{83,84} 3-MV AMS facility of the Japan Atomic Energy Research Institute,^{85,86} the AMS system at MALT (Micro Analysis Laboratory Tandem accelerator, The University of Tokyo) in Japan,^{34,86} 5-MV Uppsala tandem accelerator in Sweden,⁸⁷ and the new (operated since 2006) 1-MV multielement compact AMS facility SARA (Spanish Accelerator for Radionuclides Analyses) at the Centro Nacional de Aceleradores in Sevilla, Spain.⁸⁸ In addition, the Institute of Earth Environment of the Chinese Academy of Sciences and Xi'an Jiaotong University has jointly created an AMS center based on a 3-MV multielement accelerator mass spectrometer. This Xi'an-AMS facility has performed initial ^{129}I analyses, which will be expanded as a routine analysis.⁸⁹

5.6 Chromatographic Analyses of Inorganic Iodine Species

Chromatographic separation coupled with conductivity, spectrophotometric, or amperometric detection has been used to quantify inorganic iodine species.^{29,90–94} Hu *et al.* used a Dionex Corp. DX-600 IC system to analyze I^- and IO_3^- .²⁹ To measure low (sub-ppb) concentrations of iodide, they used pulsed amperometric detection, with a silver working electrode and an Ag/AgCl reference electrode, after iodide separation in an anion-exchange column with 50 mM nitric acid eluent. The method provides a fast (with a run time less than 3 min) and sensitive way (with a method detection limit of $0.6 \mu\text{g L}^{-1}$ at a $25 \mu\text{L}$ injection size; comparable to $0.1 \mu\text{g L}^{-1}$ by ICP-MS for total iodine analysis) to determine iodide concentration without any pretreatment. Minimal memory effect was observed, as evidenced by a measured apparent iodide concentration of only 0.005% for the first noniodide-containing sample injected immediately after a high-concentration (100 mg L^{-1}) iodide standard.²⁹

Under amperometric detection, there is no peak response for iodate. Quantitative conversion of iodate to iodide was achieved by adding 0.1 M NaHSO_3 in a volume ratio of 1 : 10 to the liquid sample, and the solution was mixed by shaking and standing for several minutes to reduce iodate to iodide; the resulting sample was analyzed for iodide by amperometry. Conversion of iodate to iodide is shown to be complete (about 100%). This approach is used to quantify the most common inorganic iodine species (iodate and iodide). In short, in addition to providing the insights on speciation of inorganic iodine, the amperometric method is also very useful in elucidating iodate–iodide interconversion.²⁹

6 SPECIATION

Iodine occurs in multiple oxidation states and as inorganic and organic species that may be hydrophilic, atmophilic, and biophilic. This section summarizes iodine speciation in air, water, and soil.

Iodine in the atmosphere exists as particle-associated, inorganic gaseous iodine (e.g., I_2 , HI, HIO), and organic gaseous iodine (CH_3I , CH_2I_2 , $\text{CH}_3\text{CH}_2\text{CH}_2\text{I}$). Hou *et al.* have summarized the speciation of ^{129}I .¹⁰ Available data, though limited, have shown that the speciation of ^{129}I in the atmosphere can supply useful information about sources and transfer pathways. Work on stable iodine species in air has been published by several authors.^{59,95–99} Organically bound iodine can be a significant fraction of total iodine in aqueous systems and in the atmosphere. Gilfedder *et al.* collected diurnal aerosol samples from Ireland and found that the majority ($90 \pm 4\%$ day samples and $94 \pm 2\%$ night ones) is soluble organically bound iodine (SOI).⁵⁹ Iodide showed higher concentrations (6% of total iodine) than iodate (1.2 of total iodine) and displayed significant enrichment

during the day compared to night. Lai *et al.* collected total suspended particle samples onboard a round-trip cruise from Shanghai, China, to Prydz Bay, Antarctica, from November 2005 to March 2006.⁹⁹ This revealed that SOI is the most abundant fraction, accounting for approximately 70% of total soluble iodine on average. Inorganic iodine species (iodate and iodide) were less than 30% of total soluble iodine. Iodide was significantly correlated with SOI, suggesting a relationship between iodide formation and SOI decomposition. Methyl iodide is an important gaseous form of iodine in the marine atmosphere and releases from nuclear-fuel reprocessing facilities, and bacteria are linked to iodine transfer from terrestrial and marine ecosystems into the atmosphere.¹⁰⁰

According to UNSCEAR, the ¹³¹I inventory in the reactor core in which the accident took place at Chernobyl was estimated to be 3.2×10^6 TBq and the total ¹³¹I released to the atmosphere over the course of the accident was about 1.7×10^6 TBq (see *Civilian Nuclear Accidents*).⁹ From four air samples collected at 14.5 m above the ground surface in Japan 10–23 days after the Chernobyl nuclear reactor accident, Noguchi and Murata reported the proportions of airborne ¹³¹I species during the Chernobyl accident: $19 \pm 9\%$ particulate iodine (iodine attached to particulate matter of mean diameter of 0.8 μm), $5 \pm 2\%$ I_2 , $6 \pm 3\%$ HIO and other non- I_2 inorganic iodine species, and $70 \pm 11\%$ organic iodine species (e.g., CH_3I).¹⁰¹ The depositional velocity of radioactive iodine from air to vegetation and water bodies and subsequent migration varied with iodine speciation. Other studies on species distribution of radioactive iodine in and around nuclear power plants and of stable iodine in the environment have also indicated the important role of organic iodine species. Aldahan *et al.* reported that the main forms of iodine released into the environment from European reprocessing facilities are alkyl iodides, CH_3I , and iodides (e.g., HI) that transform into water-soluble molecules and/or IO_3^- , which are subsequently adsorbed on and/or nucleate into aerosols and return to Earth's surface as wet or dry fallout.¹⁸

The coexistence of various inorganic and organic iodine species, in different proportions, has been reported in the hydrosphere, including in rain, snow, seawater, and groundwater.^{38,57,59,72,93,102–107} Similar to the results of iodine speciation in the atmosphere, organically bound iodine can be a significant fraction of total iodine in aqueous systems and comprises up to 50% of total iodine in aqueous samples from estuaries, rivers, and rain.⁷² SOI and iodide are the major iodine species in rain samples, with SOI accounting for 33–63% and iodide for 19–67%. However, for snow samples collected from the Black Forest in Germany, SOI is the dominant (61–75%) iodine species, followed by iodide.³⁸ In all continental rain and particularly in the snow samples, iodate is the least abundant (about <10%) species, often falling below the detection limit of about 35 ng L^{-1} .⁵⁹ In addition, iodate concentrations are always higher in coastal than inland samples, from 58 groundwater samples, Yang *et al.* found that 23 samples contained both iodide (mostly, about

80–95%) and iodate measured by IC-ICP-MS, with these inorganic iodine species comprising 84–95% of total iodine determined by ICP-MS.⁵⁷ Another 30 samples had only iodide (in the range of 15–63%), with the rest being mostly organic iodine as determined by size-exclusion chromatography-ICP-MS. The remaining five samples consisted of only iodate (no measurable iodide), in the range of 78–98% of the total iodine.

A total of 14 soil and 10 sediment samples from across the United States, along with two standard soil samples from NIST with known total iodine contents, were analyzed for iodine concentration and speciation.³⁹ These 14 soils were chosen to represent a wide range in texture, physicochemical properties, and distance from the oceans; these factors may affect the global cycling of iodine. In addition, 10 uncontaminated sediment samples were obtained from various nuclear facilities of the US. Department of Energy (DOE) where radionuclide contamination, including ¹²⁹I, could be an environmental problem. Three SRS sediment samples were collected at different depths, providing an opportunity to examine the iodine content and speciation, as well as sorption and transport of iodine species (i.e., iodide, iodate, and organoiodine in the form of 4-iodoaniline), in a vertical sediment core.

Sample properties such as pH, organic matter, texture, clay mineralogy, cation-exchange capacity, anion-exchange capacity, iron–aluminum contents extracted by both citrate–bicarbonate–dithionite and ammonium oxalate were measured according to standard procedures.³⁹ The wide range in sample types provide an opportunity to examine the correlation of total iodine content with soil properties and the influence of these properties on the transport behavior of iodine species.

Iodine species were quantified after the samples were extracted with KCl and/or TMAH solutions. A 5 mM KCl solution, with two passes, was selected to extract inorganic iodine from soil samples. Iodine extracted with 5% of TMAH at room temperature was operationally defined as the labile organic species (after subtracting inorganic iodine), and, under elevated temperature (80 °C), as total iodine. Tests with NIST SRM soils at elevated temperature produced a complete recovery of $103.9 \pm 5.1\%$ for SRM2709 and $93.8 \pm 9.4\%$ for SRM2711, respectively. About 5% of TMAH solution also extracted the inorganic iodine, and TMAH-extractable iodine was nearly identical to the combined iodine content from KCl extraction, followed by TMAH extraction.³⁹ Alkaline TMAH was first reported by Takaku *et al.* to be a favorable matrix for iodine analysis by ICP-MS.⁴³ Using TMAH for quantitative iodine extraction of soil samples was first proposed by Yamada *et al.*¹⁰⁸ and was employed in several studies.^{44,48,107}

The total iodine content in 15 US surface soil samples ranged between 0.46 and 5.42 mg kg^{-1} on a dry weight basis, where one of them, Stockton soil, is classified as a peat sample with an unusually high concentration of organic matter of 28.1%.³⁹ The average worldwide iodine content in surface soil samples (0–15 cm depth) is about 5 mg kg^{-1} .¹⁰⁹ The

iodine content in 132 UK surface soil samples differs widely based on the soil types and ranged from 0.5 to 98.2 mg kg⁻¹, with a mean of 9.2 mg kg⁻¹. The high iodine values are from soils that are close to the coast in the United Kingdom where there is relatively high rainfall and from areas with a substantial proportion of grassland with high soil organic matter. Muramatsu *et al.* reported a range of 0.2–150 mg kg⁻¹ iodine in more than 50 soil samples in Japan and high iodine concentrations are likely directly influenced by the marine atmosphere, high rainfall (about 2 m per year) and high adsorption capacity of iodine on Andosol soils in Japan.¹¹⁰

In addition to measurements of the total iodine content, Hu *et al.* assessed iodine speciation in soils and sediments.³⁹ In natural soil samples, iodine is mostly (nearly 90% of total iodine) present as organic species, among them appreciable amounts (about 50% for most soils) are nonlabile, i.e., extractable with 5% TMAH under only elevated temperature (Figure 2). In contrast, inorganic iodine becomes important (up to 50%) in sediments with low organic matter; however, organic species are still the dominant form of iodine in these sediments.

Inorganic iodine, extractable with a low-ion-strength KCl, comprised less than 10% of the total iodine in the surface soils. This is consistent with the reports that approximately 80% of 183 UK soils contained less than 10% cold-water extractable iodine.¹ For some soil samples, the proportion of iodide and iodate in inorganic iodine was also evaluated by Hu *et al.*, and it seems that either may be dominant.³⁹ There are very limited reports on the distribution of iodide and iodate in soil iodine, partly because of the limitation of analytical approaches sensitive enough to evaluate their speciation in soils. Yamada *et al.* assumed that iodide is more soluble

than iodate and assigned the first two repeats of extraction with 0.1 M KCl as the iodide content and the subsequent two extractions as the iodate content.¹⁰⁷ They reported that iodide and iodate contributed about equal amounts (about 5 mg kg⁻¹) in a red-yellow mountain soil, while iodate (about 6 mg kg⁻¹) was the only inorganic iodine found in another Andosol virgin soil.

Regression analyses of various iodine components (inorganic, labile organic, nonlabile organic, and total iodine) with different physicochemical properties of surface soils were reported by Hu *et al.*³⁹ The correlation is highest for organic matter and cation-exchange capacity, followed by CBD-extractable Al, clay content, and CBD-extractable Fe. Overall, it confirms the dominant influence of organic matter, clays, and sesquioxides in retaining iodine in soils. The good correlation between iodine and organic matter in surface soils is likely related to the supply (atmospheric input) of iodine and its retainability.^{1,111} However, in 154 samples collected in successive 10-cm increments from 18 soil profiles in the United Kingdom, the total iodine content was closely correlated with oxalate-soluble Al but not with oxalate-soluble Fe or organic matter.¹¹² The parent rock material from which soils are derived can also influence soil iodine concentrations, with organic-rich shales and carbonates at the high end of the range and granites and basalts at the low end.^{37,113}

7 GEOCHEMICAL BEHAVIOR

Several iodine species exist in earth reservoirs and are involved in the cycling of iodine in the global environment.

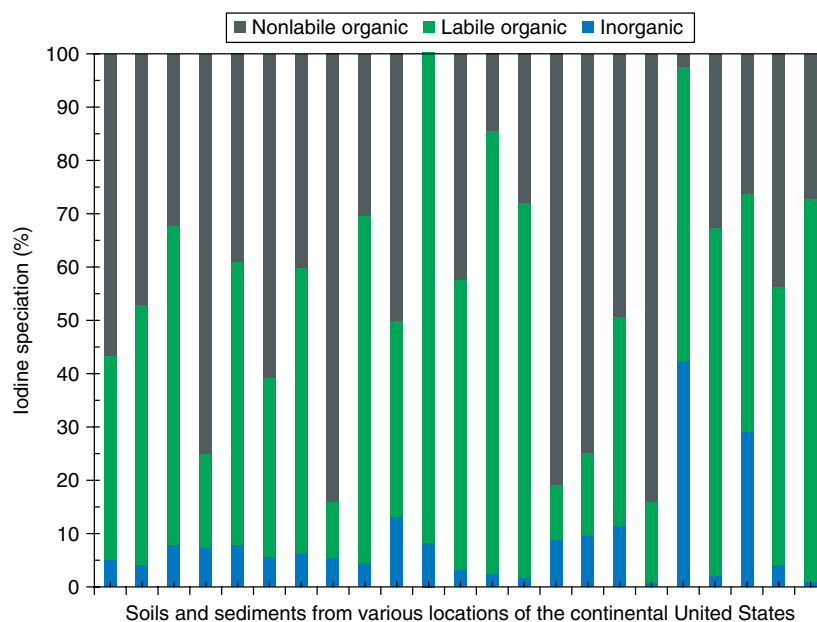


Figure 2 Iodine speciation in soils and sediments from various locations in the continental United States

Various physicochemical properties in the subsurface will affect sorption and transport behavior of iodine species. Hu *et al.* studied the transport of iodine species (iodide, iodate, and 4-iodoaniline), along with tracers of bromide and tritium, in various soils and sediments from DOE nuclear operation facilities.^{29,39} A very limited sorption of I^- during transport in many samples (including SRS surface soil, SRS aquifer, and Hanford sediment samples) was observed, which is consistent with other reports of negligible sorption of I^- .^{114,115} However, soil properties play an important role in the sorption of I^- . There was significantly retarded transport of I^- , compared to bromide/tritium, in the SRS subsoil sample. This sample contained an appreciable amount of iron and aluminum oxide minerals, which possesses positively charged surfaces that contribute to anionic I^- sorption.

Iodate commonly exhibits more retardation during transport than iodide because it interacts more strongly with both clays and organic matter.^{104,116–118} As discussed by Kaplan *et al.*,¹¹⁵ the cause for the difference in I^- and IO_3^- sorptive behavior is not known but is presumably a result of the harder base nature of IO_3^- , as compared to I^- , which would favor hard-hard interactions with the hard acid sites on the mineral surfaces.¹¹⁵ The disparate sorption behavior of I^- and IO_3^- was observed in transport studies in several soils and sediments, which consistently showed greater sorption of IO_3^- than for I^- . Even in Hanford sediment, which has a very low anion-exchange capacity, notable sorption of IO_3^- took place.²⁹

Using amperometric detection, Hu *et al.* found that a portion of iodate, after being in contact with several soils (Hanford and SRS sediments) and clay minerals (kaolinite, illite, and montmorillonite), was converted to iodide, and this abiotic reduction was probably mediated by structural iron (Fe) present in the clay minerals.²⁹ Some of the most abundant natural reductants are soil clay minerals that contain reduced forms of iron and sulfur. Using X-ray absorption near edge structure, which requires a relatively high concentration (0.009 M iodine as IO_3^-), Furrmann *et al.* reported that IO_3^- was reduced to what appeared to be I_2 or HI vapor in the presence of pyrite; no change in the iodine oxidation state was observed in magnetite and biotite experiments.¹¹⁹ Fukui *et al.* found that I^- accounted for about 30% total iodine, with IO_3^- as the starting species, in a solution after a 14-day contact with a Japanese soil; no explanation for the reduction was offered.¹²⁰ Muramatsu *et al.* reported that IO_3^- was mostly (68–95%) converted to I^- after an 8-day contact with three Japanese soils, and there was a negligible change for I^- .¹²¹

Compared to I^- and IO_3^- , studies regarding transport of organoiodine compounds are much more scarce, given the challenges associated with the analysis of various organoiodine compounds. Hu *et al.* used 4-iodoaniline as a representative nonvolatile organoiodine species and found that the transport of 4-iodoaniline is quite different from that of the inorganic forms, I^- and IO_3^- , and is related to the

amount of organic matter in the sample.^{29,39} This finding is consistent with the hydrophobicity of organoiodine and its affinity for hydrophobic organic matter. In SRS surface soil with 2.33% of organic matter, transport as 4-iodoaniline is delayed by more than 20 times than that of I^- . In contrast, 4-iodoaniline migrates without retardation, similar to bromide and tritium, in the SRS subsoil, while IO_3^- and I^- are significantly retarded because of the strong anion-exchange capacity in this sample. This is also true for the SRS aquifer material, though the magnitude of sorption for IO_3^- and I^- is less than that for SRS subsoil. Disparate transport behavior of three different iodine species, in three types of samples in a vertical cross section from the SRS, highlights the need to consider sediment properties when predicting the geochemical behavior and transport of iodine.

The input concentration and the contact time also have an effect on iodine sorption and transport behavior. Because the sorption of both I^- (especially) and IO_3^- are related to the presence and the magnitude of soil components that possess positively charged surfaces and since positively charged surface sites are limited, the extent of sorption is expected to be affected by the input concentration and by the presence of competing ions. Using an input concentration of 10^{-5} – 10^{-4} M, Hu *et al.* studied the sorption and transport of I^- and IO_3^- in SRS and Hanford sediments.²⁹ The trend of I^- and IO_3^- sorption with respect to the initial concentration is consistent with the limited number of positively charged sites for sorption. Greater I^- sorption at a much lower (10^{-12} M) concentration for three subsurface sediments from the Hanford Site was reported by Kaplan *et al.*¹¹⁵

Ideally, one would use a concentration that is comparable to real-world situations, i.e., 10^{-8} M for stable iodine in the hydrosphere¹ and a few orders of magnitude lower for anthropogenic ^{129}I , as observed at SRS.¹²² However, for trace concentrations of I^- , usually introduced as radioactive ^{125}I in laboratory experiments, in a system open to air, the I^- can be readily oxidized to IO_3^- , which complicates data interpretation, considering the different sorption behavior of I^- and IO_3^- discussed above.^{104,119} It is not improbable that observed sorption of iodide, described in some studies, is actually the result of conversion to IO_3^- and subsequent “apparently higher” sorption.

The effect of residence time on biogeochemical cycling of iodine also needs to be considered; this is more critical for anthropogenic ^{129}I , which has only been involved in iodine cycling over past 50 years. Because of the longer interaction times (thousands of years) and the higher concentrations of ^{127}I , compared to ^{129}I , ^{127}I preferentially occupies the thermodynamically favorable sorption sites. The relative portion of reversible binding sites is substantially higher for ^{129}I than for ^{127}I on the time scale of anthropogenic ^{129}I fallout.²⁸ Therefore, trace ^{129}I possesses a higher mobility than stable ^{127}I .²⁷ Overall, anthropogenic ^{129}I , with its lower concentration and short contact time with soils, will likely take a prominent role in the geochemical cycle; the

presence of different iodine species in the anthropogenic ^{129}I input further complicates the biogeochemical cycling of ^{129}I .

8 POTENTIAL HUMAN EXPOSURE

Eisenbud and Gesell reported that 93 TBq of ^{129}I was produced by the nuclear power industry up to the year 2000.¹²³ Over the time period 1956–1988, the PUREX plant at Hanford processed more fuel than all the other Hanford separation plants combined. With a cumulative release of 0.873 TBq ^{129}I , the PUREX plant was responsible for the majority of ^{129}I emissions at the Hanford Site; more than 90% of the total ^{129}I was present as vapor-phase material.¹²⁴ From the monitoring of ^{129}I activity in air and milk samples, atmospheric emissions were found to be the major source of ^{129}I in environmental samples.

Iodine ingested from food and inhaled from the air will be transferred into blood, with most of the iodine uptake concentrated in the thyroid, the target organ for iodine in mammals. The thyroid takes up stable and radioactive iodine indiscriminately.¹⁰ Because of its low β - (and long-life life) and γ -energy, ^{129}I only poses a long-term and very low-dose exposure. Hou *et al.* reported that, for an $^{129}\text{I}/^{127}\text{I}$ ratio of 10^{-6} , the effective radiation dose to the thyroid from the internal exposure of ^{129}I is less than 10^{-3} mSv per year, which is 40 times lower than 0.04 mSv per year, the maximum contaminant level for “combined β - and photon emitting radionuclides” in drinking water.¹⁰ For the highest $^{129}\text{I}/^{127}\text{I}$ ratio of 10^{-4} observed in areas close to nuclear reprocessing facilities, the annual radiation dose could be 0.01 mSv year. These calculations do not consider the uptake of stable iodine from the diet with very low ^{129}I concentrations.

For the intake of radionuclides with stable isotopes, such as ^{129}I and ^{14}C , the concentration of the stable isotope should be considered in the dose calculation.^{125,126} Considering the stable iodine concentration in the groundwater near Yucca Mountain, the potential geological repository for storing high-level nuclear wastes in the United States, yields an upper bound on the annual committed thyroid dose of 1.2×10^{-5} mSv (1.2×10^{-3} mrem). When the additional intake of stable and radioactive iodine in other components of the diet is considered, the annual committed effective dose would be about 2.5×10^{-8} mSv (2.5×10^{-6} mrem), less than 10^{-6} of the Individual Protection Standard of 15 mrem.

9 CONCLUSIONS

This article summarized the basic chemistry, occurrence, detection methods, speciation, and geochemical

behavior of iodine and radioiodine, taking into consideration the similarities and differences between the radionuclides and stable iodine. The following can be summarized regarding the current understanding and suggested future work on iodine in the environment.

- Redox-sensitive iodine occurs as multiple species (inorganic and organic) in the environment, each with varying hydrophilic, atmophilic, and biophilic characteristics.
- Various iodine species in different proportions, therefore, exist in the atmosphere, hydrosphere, and lithosphere. Organically bound iodine, with limited solubility and mobility, commonly comprises the major portion of total iodine in soil and sediment, which is commonly correlated with total organic matter, sesquioxides, and clay content.
- Different iodine species exhibit very different sorption and transport behavior in the subsurface. Sorption of iodate is consistently greater than that of iodide, while transport of organoiodine is quite limited and related to the amount of organic matter in the matrix. The physical and chemical processes affecting iodine transport include iodate reduction, irreversible retention or mass loss, rate-limited and nonlinear sorption of iodine, and interconversion of iodine species, both abiotically and biologically.
- Examination of iodine speciation, with due attention to potential interconversion among species, is essential when interpreting the environmental behavior of iodine. Conflicting reports of iodine cycling in the environment could be due to the lack of understanding of iodine speciation and their disparate environmental behavior.
- With a shorter contact time than stable iodine, anthropogenic ^{129}I will likely have a higher mobility. However, ^{129}I will experience a similar speciation process as stable iodine and may eventually be retained strongly, with organoiodine as the dominant species.
- Coupled with a mature set of extraction techniques, hyphenated separation (e.g., chromatography) and detection (ICP-MS, amperometry) have proven to be versatile methods to study stable iodine, while AMS is the method of choice for analyzing ^{129}I . However, further development of sensitive and specific analytical methods for different iodine species in complex matrices is warranted. Multiple and integrated approaches are needed to tackle complex issues surrounding iodine speciation and biogeochemical cycling in the environment.

10 RELATED ARTICLES

Anthropogenic Radioactivity; Natural Radioactivity; Oceans and Seas; Speciation.

11 ABBREVIATIONS AND ACRONYMS

AMS = accelerator mass spectrometry; DOE = Department of Energy; IC = ion chromatography; ICP-MS = inductively coupled plasma-mass spectrometry; LSC = liquid scintillation counting; NAA = neutron activation analysis; NIST SRM = National Institute of Standards and Technology Standard Reference Material; NSF = National Science Foundation; NSRL = Nuclear Structure Research Laboratory; SOI = soluble organically bound iodine; SRS = Savannah River Site; TMAH = tetramethyl ammonium hydroxide.

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