Analysis of iodine-129 in environmental materials: Quality assurance and applications

S. Szidat,[1]* A. Schmidt,[1] J. Handl,[1] D. Jakob,[1] R. Michel,[1] H.-A. Synal,[2] M. Suter[3]

[1] Zentrum für Strahlenschutz und Radioökologie, Universität Hannover, Am Kleinen Felde 30, D-30167 Hannover, Germany
[2] Paul Scherrer Institut, c/o Institut für Teilchenphysik, ETH Hönggerberg, CH-8093 Zürich, Switzerland
[3] Institut für Teilchenphysik, ETH Hönggerberg, CH-8093 Zürich, Switzerland

The long-lived radionuclide ^{129}I ($T_{1/2} = 15.7$ My) occurs in the nature in very low concentrations. Since the middle of our century the environmental levels of ^{129}I have been dramatically changed as a consequence of civil and military use of nuclear fission. Its investigation in environmental materials is of interest for environmental surveillance, retrospective dosimetry and for the use as a natural and man-made tracers of environmental processes. We are comparing two analytical methods which presently are capable of determining ^{129}I in environmental materials, namely radiochemical neutron activation analysis (RNAA) and accelerator mass spectrometry (AMS). Emphasis is laid upon the quality control and detection capabilities for the analysis of ^{129}I in environmental materials. Some applications are discussed.

Introduction

The environmental abundances of the long-lived radionuclide ^{129}I ($T_{1/2} = 15.7$ My) have been sustainably changed by man due to atmospheric nuclear weapon tests and, even more, due to emissions from nuclear reprocessing plants. Though its present concentrations in the biosphere do not produce any significant radiation exposure, the development of future ^{129}I levels in nature have to be carefully monitored. Moreover, ^{129}I is a versatile and important natural and anthropogenic radioactive tracer to investigate large scale environmental processes. For both environmental monitoring of ^{129}I and for its application as tracer, the knowledge about its natural, prenuclear levels is essential. This knowledge is, however, still incomplete and a lot of contradictory data are found in the literature.[1]

Practical applications of ^{129}I include the retrospective dosimetry of the ^{131}I exposition of people living in the vicinity of Chernobyl long after the short-lived ^{131}I decayed[2,3] and the documentation of the present status and the surveillance of future sites for the final disposal of radioactive waste.

There are two analytical techniques which are sensitive enough to investigate ^{129}I in environmental materials, namely the radiochemical neutron activation analysis (RNAA)[4,5] and the accelerator mass spectrometry (AMS).[6,7] However, as will be shown below, only AMS is capable of covering the natural, prenuclear levels and much of the incomplete knowledge about the prenuclear levels has to be attributed to the lack of quality assurance in older RNAA work.

Therefore, we present here a critical comparison of the analytical capabilities of both methods and discuss in some detail the required procedures of quality assurance.

Experimental

In 1962, RNAA was used[4] for the first time to determine ^{129}I and stable ^{127}I. It was for many years the only method capable of measuring ^{129}I in environmental samples and a large number of investigations was performed mainly by monitoring the elevated ^{129}I levels from atmospheric weapon tests and from releases of nuclear installations. Since 1980, ^{129}I could also be determined via AMS[6] and because of its outstanding sensitivity it caused a revival of ^{129}I analyses and manifold applications.[7]

Since ^{129}I is always accompanied in nature by ubiquitous stable iodine it is necessary to discuss ^{129}I abundances not only in terms of their concentrations but also as ^{129}I/^{127}I isotopic ratios. This is also of importance with respect to the modeling of the radiological consequences of ^{129}I in the environment. The advantage of RNAA is that it is capable to determine both isotopes ^{129}I and ^{127}I simultaneously in one sample. Since via AMS only isotopic ratios are determined, a second analytical technique such as ICP-MS or ion-chromatography (IC) has to be applied to determine stable ^{127}I.

In any case, ^{129}I and ^{127}I have to be extracted from the sample material prior to their analysis and a variety of separation schemes has to be used depending on the matrices under investigation.

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* E-mail: michel@mbox.zsr.uni-hannover.de

For RNAA of soils, tissues and vegetation, iodine is usually separated from the sample matrix by combustion techniques (described below) as I_2 and collected on charcoal. After purification by distillation, the iodine is irradiated in a sealed quartz ampoule for 2 hours with appropriate standards, in our case, in the FRG-1 research reactor of GKSS at Geesthacht/Germany at a thermal neutron flux of $4.4 \cdot 10^{13}$ n·cm^{-2}·s^{-1}. The epithermal and fast fluxes in this position are $1.3 \cdot 10^{12}$ and $5.7 \cdot 10^{12}$ n·cm^{-2}·s^{-1}, respectively. After post-irradiation chemistry to separate from other radioactive substances, in particular activated bromine isotopes, the iodine is precipitated as AgI and measured by γ-spectrometry for ^{125}I (as tracer for the determination of the chemical yield), ^{126}I and ^{130}I via their 35.5 keV, 389 keV and 536 keV γ-energies, respectively. For further details see, e.g., Reference 8.

For AMS measurement, the extracted iodine has to be transformed into at least 1 mg of silver iodide of which the $^{129}I/^{127}I$ ratio is measured by AMS. Our AMS measurements are performed at the PSI/ETH Tandem AMS facility of ETH Hönggerberg at Zurich/Switzerland, (See References 7 and 9 for details of the AMS measurement). Since most environmental materials do not contain enough intrinsic iodine to extract from a convenient sample mass enough for 1 mg AgI, it is necessary to add some iodine carrier which has a low ^{129}I content, so-called Woodward iodine from Woodward Corporation, Oklahoma, USA with $^{129}I/^{127}I = 4 \cdot 10^{-14}$.[10] This carrier addition can be done in two different ways:

(a) In the so-called "carrier method", the stable iodine carrier is added to the sample prior to extraction of the iodine. If the sample material has negligible intrinsic stable iodine and if the amount of carrier which has been added to the sample is known, the absolute ^{129}I concentration of the sample can be calculated from the $^{129}I/^{127}I$ ratio measured by AMS. The chemical yield of the extraction can be determined from the recovery of the stable iodine carrier.

(b) In the so-called "tracer method", the stable iodine carrier is added after extraction of ^{129}I and intrinsic ^{127}I from the sample before precipitation of AgI. The intrinsic ^{127}I content can be determined by IC or ICP-MS from an aliquot of the extracted iodine taken prior to carrier addition. ^{129}I concentration is calculated from the $^{129}I/^{127}I$ ratio measured by AMS by multiplication with the total ^{127}I in the AMS sample. The $^{129}I/^{127}I$ ratio of the sample material is derived from the ^{129}I and intrinsic ^{127}I concentrations. In this case ^{125}I must be used as a radioactive tracer to determine the yield of the steps prior to carrier addition. For further details see Reference 1.

The following separation techniques are used to extract iodine from environmental samples:

(a) Up to 70 g soil, 7 g vegetation or 3 g soft tissue samples are combusted in a stream of oxygen at a temperature of 1100 °C after a careful increasing of the temperature with a Bunsen burner ("combustion method"). For RNAA the generated gaseous iodine is trapped on activated charcoal, for AMS it is reduced to iodide in an alkaline $NaHSO_3$ solution.[1]

(b) Alternatively, 2 g soil, vegetation or soft tissue samples can be leached in a nickel crucible with sodium hydroxide after adding iodide and iodate (Fluka puriss., $^{129}I/^{127}I = 2 \cdot 10^{-13}$) carriers for 1 hour at 150 °C, then for 2 hours at 200 °C followed by 3 hours at 275 °C. The melt is extracted into water and reduced by 0.1 mol·l^{-1} $NaHSO_3$ ("alkaline leaching method"). After acidification with HNO_3 and oxidation with 1 mol·l^{-1} $NaNO_2$ iodine is extracted into chloroform; back-extraction into water is done under reducing conditions. This last step is repeated twice.[1]

(c) Aerosol samples are suspended with the filter in an alkaline $NaHSO_3$ solution. The mixture is heated at the boiling point for 1 hour and filtered after cooling down. Purification steps by extraction and back-extraction as described above are carried out.[11]

(d) From water samples (surface water, ground water or precipitation), iodide is extracted by BIORAD AG 1 anion exchange resin after an oxidation-reduction step[12] to convert all inorganic iodine species into iodide. The analyte is eluted with 2 mol·l^{-1} $NaNO_3$.[1] Fresh and homogenized milk can be treated in the same manner with no preceding oxidation-reduction step.

Results and discussion

Detection capabilities

The capabilities to determine ^{129}I by RNAA are mainly downwards limited by the γ-spectrometric background of the ^{130}I measurement which itself depends on the quality of purification from bromine in the post-irradiation chemistry and on the intrinsic content of ^{127}I. For AMS, ^{129}I blank concentration of the carrier iodine and of the AMS measurement are the limiting factors. In Table 1, we compare the lowest detection limits[13] of RNAA and AMS achieved in the analysis of ^{129}I in thyroid glands, soil samples and plant materials. In addition, we give the ranges of lowest $^{129}I/^{127}I$ ratios which can be determined. They depend on the detection limits of ^{129}I and on the ranges of ^{127}I amounts in the maximum sample masses which can be handled in one analysis.

Table 1. Lowest detection limits according to DIN 25482-6 (ISO 11929-2)[13] achieved in the determination of ^{129}I by RNAA and AMS and range of lowest ^{129}I/^{127}I isotopic ratios which can be determined

Material	AMS			RNAA		
	^{129}I, pg·kg^{-1}	^{129}I, µBq·kg^{-1}	^{129}I/^{127}I, ×10^{-10}	^{129}I, pg·kg^{-1}	^{129}I, µBq·kg^{-1}	^{129}I/^{127}I, ×10^{-10}
Thyroids	0.46	3.0	0.0015–0.015	2,000	13,000	6.4–64
Soil	0.023	0.15	0.0075–0.45	125	810	4.1–250
Plants	0.20	1.3	0.5–39	840	5,500	2,000–170,000

On the average, the detection limits of AMS are about 3 orders of magnitude lower than those of RNAA. The same is true for the accessible ^{129}I/^{127}I ratios which in case of RNAA all are above $4 \cdot 10^{-10}$. This fact causes a big problem. The prenuclear, natural equilibrium ratio in the environment was estimated to be ^{129}I/^{127}I $= (0.3–3) \cdot 10^{-12}$.[14] This estimate is well in agreement with recent AMS measurements of young oceanic sediments which yielded ^{129}I/^{127}I $= (1.3 \pm 0.3) \cdot 10^{-12}$.[15] Today, atmosphere, hydrosphere, pedosphere and biosphere are sustainable polluted by man-made ^{129}I[1] for a survey of available data and a detailed discussion. When ^{129}I analyses of environmental analyses started in the 1960s, the natural ratios were already changed and, more important, RNAA was not capable to analyze prenuclear ratios even if appropriate sample material would have been found. There are two orders of magnitude of ^{129}I/^{127}I ratios between the natural levels and the detection limits of RNAA. This difference leaves large white areas on the environmental ^{129}I map. Thus, information about the natural, prenuclear ^{129}I in the environment and about its transition into modern elevated levels can presently only be obtained by AMS.

Quality control

Quality assurance of ^{129}I analyses has for long been and still is a problem. In the older literature, neither reports about laboratory blanks, which were simply not accessible by RNAA, nor about measures of quality control are to be found. We discuss here shortly some aspects of the current situation.

The quality assurance of ^{129}I and ^{127}I analyses has to comprise different aspects, namely control of blanks, of accuracy and of the analytical techniques used. Blank control means that each sample treatment procedure must be verified with respect to blank contents of chemicals used, contamination during separation processes and memory effects of laboratory ware. The validity of the analytical techniques has to be proven by analyses using different independent techniques, here RNAA and AMS. Accuracy control has to verify that intrinsic ^{129}I and ^{127}I and carriers are equilibrated and

behave in the same way during the various separation steps and, finally, that certified data of standard reference materials can be reproduced.

With respect to blank control, a severe contamination problem occurred when we started AMS analyses of low-level samples after twenty years of RNAA of high-level ^{129}I materials. As described elsewhere in detail,[1] up to 3 µBq (0.5 pg) ^{129}I were brought into each analysis via chemicals and laboratory ware. The resulting blank values did not affect the measurements with RNAA but those with AMS with its much lower detection limit. Iodine can occur in many different highly volatile compounds and is easily absorbed by laboratory materials. We could not solve this problem by simply changing laboratory ware. Setting-up of a new low-level laboratory without any iodine history was necessary. From that time on, samples of expectedly low or high ^{129}I contents were prepared in the respective laboratory. This led to blank values in the low-level laboratory lower than the blank values of the AMS measurements themselves which are determined by repeated measurement of Woodward iodine. However, significantly raised laboratory blanks are still occasionally observed making blank control a permanent task. Causes for this were, e.g., contaminated $NaNO_3$ chemicals, used sample containers and combustion glassware or a faulty water purification system.

Accuracy checks of ^{129}I analyses and comparison of RNAA and AMS are possible by analysis of the only reference material (RM) existing for ^{129}I, i.e., IAEA soil 375.[16] This RM is a soil from the vicinity of Chernobyl which was certified by a round robin test of several RNAA laboratories and which has a high ^{129}I/^{127}I ratio of approx. $1.5 \cdot 10^{-7}$.[17,18] We investigated this RM by AMS and IC after carrier combustion, tracer combustion and alkaline leaching and by RNAA after tracer combustion (Table 2). Tracer combustion is the only method that allows determination of ^{127}I content and thus of the intrinsic ^{129}I/^{127}I ratio of the sample; carrier combustion uses excess iodine carrier before combustion and is so nearly unaffected by the chemical yield. Both analytical techniques and all sample preparation methods reproduced the certified value within statistical errors and thus proved the accuracy of both, AMS and RNAA for high-level soil samples.

Table 2. Concentrations (relative to dry weight) of ^{129}I and ^{127}I with standard uncertainties in IAEA soil 375 determined by RNAA and by combination of AMS and IC. The standard uncertainty of the reference value was calculated from the given confidence interval

Procedure	n	^{129}I, pg·kg^{-1}	^{129}I, µBq·kg^{-1}	^{127}I, mg·kg^{-1}	^{129}I/^{127}I, ×10^{-10}
Tracer combustion (AMS/IC)*	3	268 ± 11	1750 ± 70	1.70 ± 0.04	1550 ± 40
Carrier combustion (AMS)	2	265 ± 4	1730 ± 30	–	–
Alkaline leaching (AMS)	1	270	1760	–	–
Tracer combustion (RNAA)	4	250 ± 70	1600 ± 500	1.68 ± 0.09	1500 ± 500
IAEA reference value[16]	10	262 ± 29	1710 ± 190	(2)[17]	~1300

* Results of one analysis were already reported earlier.[1]

Table 3. Concentrations (relative to dry weight) of ^{129}I and ^{127}I with standard uncertainties in NIST SRM 2709 (San Joaquin soil) determined by combination of AMS and IC after tracer combustion

Source of data	^{129}I, pg·kg^{-1}	^{129}I, µBq·kg^{-1}	^{127}I, mg·kg^{-1}	^{129}I/^{127}I, ×10^{-10}
This work, mean*	1.0 ± 0.4 (n = 2)	6.5 ± 2.6 (n = 2)	4.7 ± 0.5 (n = 3)	2.3 ± 1.1 (n = 2)
NIST informative[22]	–	–	5	–
Marchetti et al.[18]	–	–	4.67 ± 0.32 (n = 5)	–

* Results of one analysis were already reported earlier.[1]

For low-level samples neither a second analytical technique except AMS exists presently nor are there any RMs. The latter poses also a problem for the AMS analysis of ^{129}I as it became evident in a recent round robin test.[19] The pure AMS measurements of ^{129}I in AgI and water samples agreed well within the uncertainties of the individual measurements. However, the results of biospheric samples as pine needles, maple leaves, seaweed and soil differed by nearly three orders of magnitude making sample preparation and insufficient blank control the likely cause of these discrepancies. Therefore, in a second phase of this round robin,[20] sample preparation was performed by some selected laboratories and by measurements of all facilities which participated in the first phase. Now, results for the high-level IAEA soil 375 differed only by a factor of two with a mean matching the reference value. Results for a low-level maple leaf sample still spread over more than one order of magnitude.

These experiences call urgently for the establishment of RMs with small ^{129}I content and ^{129}I/^{127}I ratios. From our own analyses, we propose SRM NIST 2709 (San Joaquin Soil, baseline trace elements) with an isotopic ^{129}I/^{127}I ratio of ~2.3·10^{-10} as a suitable low-level RM for the analysis of ^{129}I (Table 3). The further open problem of quality assurance of ^{127}I determination via IC or ICP-MS and the lack of suitable RMs is not discussed here.

Applications

Our present investigations of ^{129}I in environmental materials have three goals: (a) the establishment of reliable prenuclear ^{129}I levels and ^{129}I/^{127}I ratios for all relevant environmental compartments as a data base for the assessment of the long-term human impact on nature, (b) the description of the actual radioecological status of the environment to allow for a detailed balance of ^{129}I in the vicinity of nuclear waste depositiories and other nuclear installations, and (c) the measurement of ^{129}I in Ukrainian soils in areas highly contaminated by Chernobyl fallout for sake of retrospective dosimetry of ^{131}I exposure.

Here, we shall just give two examples dealing with the prenuclear levels and the signal of the Chernobyl accident in soils which both are essential with respect to the rating of RNAA and AMS capabilities.

The first example deals with the problem of prenuclear biospheric ^{129}I/^{127}I ratios which according to earlier estimates should match the natural equilibrium ratio of ~10^{-12} in the hydrosphere. An AMS analysis in our laboratory of a thyroid gland powder from USA with a manufacturing data of Febr. 1943 (Parke-Davis, #C547B) yielded a value of (7.0±1.5)·10^{-12} which is much lower than older literature data[1] pointing to severe blank and sensitivity problems in the earlier RNAA analyses, but still being higher than the expected equilibrium ratio.

Table 4. Concentrations (relative to dry weight) of ^{129}I and ^{127}I in various soils determined
by combination of AMS and IC after tracer combustion

Material	^{129}I, pg·kg^{-1}	^{129}I, μBq·kg^{-1}	^{127}I, mg·kg^{-1}	^{129}I/^{127}I, ×10^{-10}
Moscow, 1910[a]	0.113	0.74	0.77	1.4
Moscow VI, 1996[a,b]	1.2–102	7.8–669	1.2–2.9	9.8–403
Nemirowka II, 1995[a,b]	1.2–339	7.5–2,216	4.5–7.9	2.5–526
Naroditschi II, 1995[a,b]	9.3–783	60.8–5,118	0.5–0.72	187–13,110
Lower Saxony, 1998	70–153	460–1,000	0.7	1,000–2,100

[a] Range of data measured in 40 cm soil profiles.
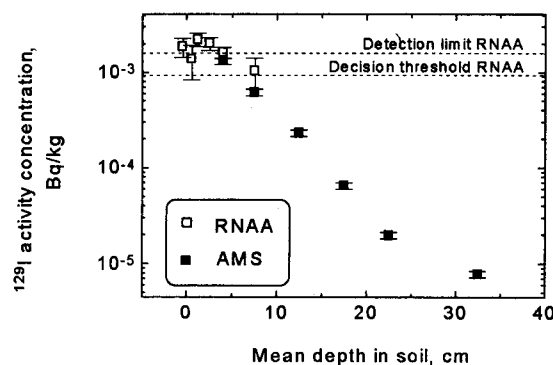[b] For some details see Ref. 3.

Fig. 1. ^{129}I in the soil profile Nemirowka II as measured by RNAA and
AMS. The actual detection limit and decision threshold of RNAA
according to DIN 25482-6 (ISO 11929-2)[13] are indicated

The second example is a comparison of ^{129}I and ^{127}I
analyses in various samples of top-soils and soil profiles
from Moscow, Ukraine and Lower Saxony/Germany
(Table 4). In the analysis of the 40 cm soil profile
Nemirovka II from contamination zone III[21] around
Chernobyl RNAA turned out to be only capable to
analyze samples from the topmost 10 cm (Fig. 1).
Deeper samples were accessible by AMS, only.
Maximum activities in the profiles Naroditschi II
(contamination zone II)[21] and Nemirovka II are
significantly higher than from Moscow VI and of top-
soils from Lower Saxony which did not suffer from large
fallout from the Chernobyl accident. A detailed
discussion of the data showed that the fallout due to
Chernobyl can be reliably determined from the total
deposition densities measured in the Ukrainian
samples.[1,3] The total deposition densities of ^{129}I which
provide the basis for retrospective dosimetry of ^{131}I
exposure has to be determined either by integrating over
measured ^{129}I depth profiles or, more economical, by
measuring mixed samples from entire profiles. In both
cases, exclusively AMS is capable of measuring ^{129}I in
all samples needed.

Finally, the question of ^{129}I concentrations and
^{129}I/^{127}I ratios in prenuclear soils shall be addressed. In
a soil profile from Moscow taken in 1910, ^{129}I could

only be measured by AMS analyzing a combined
sample. The analysis resulted in a ten times lower ^{129}I
concentration than found in all other samples with a
^{129}I/^{127}I ratio of only 1.4·10^{-10}. It is still not clear
whether these numbers really reflect prenuclear levels in
soils or whether they are affected by contamination
during a many-decades-long storage. Therefore, more
work remains to be done, AMS being the proper nuclear
analytical technique for such investigations.

In spite of the much higher detection capabilities of
AMS, one can, however, conclude that both RNAA and
AMS provide valuable tools for the analysis of ^{129}I in
the environment, provided that the two methods are used
with strict protocols of quality assurance and that the
individual detection capabilities are taken properly into
account.

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