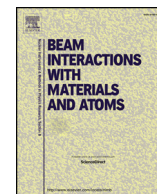




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## Nuclear Inst. and Methods in Physics Research B

journal homepage: [www.elsevier.com/locate/nimb](http://www.elsevier.com/locate/nimb)Independent measurements of  $^{129}\text{I}$  content in environmental reference materials using accelerator and thermal ionization mass spectrometryJ.E. Olson<sup>a,\*</sup>, M.L. Adamic<sup>a</sup>, D.C. Snyder<sup>a</sup>, J.L. Brookhart<sup>a</sup>, P.A. Hahn<sup>a</sup>, M.G. Watrous<sup>a</sup><sup>a</sup> Idaho National Laboratory, P.O. Box 1625, Idaho Falls, ID 83415, USA

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## ABSTRACT

Iodine environmental measurement programs are in need of new materials with certified  $^{129}\text{I}$  activity. Frequently  $^{129}\text{I}$  measurements are validated in the literature using the standard material IAEA-375, Chernobyl soil, which is the only soil/sediment material with a recommended  $^{129}\text{I}$  activity. IAEA-375 has not been available for purchase since 2010. This study is an extension of previous work at INL to include four additional standard materials that are commercially available (NIST materials: RM 8704, Buffalo River sediment, SRM 2710a, Montana I soil, and IAEA materials: SL-1, lake sediment, IAEA-385, Irish Sea sediment). These materials have certified or recommended activities for a variety of radionuclides but not for  $^{129}\text{I}$ . This paper reports a comparison of accelerator mass spectrometry (AMS) and thermal ionization mass spectrometry (TIMS) data for  $^{129}\text{I}$  activity, as well as the  $^{129}/^{127}\text{I}$  ratios for these standards to assist in identifying a suitable alternative for IAEA-375. Two independent chemical separation and mass spectrometric analysis techniques have been applied in an effort to corroborate the data. Both methods were validated via analyses of IAEA-375 for  $^{129}\text{I}$  and show good agreement with the recommended activity of  $1.7 \times 10^{-3} \text{ Bq kg}^{-1}$  for  $^{129}\text{I}$ ; ( $1.6 \times 10^{-3} \text{ Bq kg}^{-1}$  by AMS and  $1.8 \times 10^{-3} \text{ Bq kg}^{-1}$  by TIMS) with both sets of results within the 95% confidence interval of the recommended value.

## 1. Introduction

Due to its long half-life ( $t_{1/2} = 15.7 \text{ Ma}$ ),  $^{129}\text{I}$  has proven to be very useful as a tracer for environmental processes and age dating up to 80 million years [1,2] leading to a growing number of AMS laboratories around the world analyzing  $^{129}\text{I}$ . Along with new programs and as more innovative approaches to iodine processing are being developed, comes an increase in the demand for readily available standards with known concentrations of iodine. Iodine environmental measurements on soil or aerosol samples have consistently been validated in the literature by using the standard material IAEA-375, soil from Novozhybkov, Russia, a city roughly 260 km northeast of Chernobyl, which is the only soil/sediment material with a recommended  $^{129}\text{I}$  activity [3,4,5,6,7]. IAEA-375 has not been available for purchase since 2010.

A number of other environmental standards have been either certified or analyzed for  $^{129}\text{I}$  concentration; however, most of these are also no longer available [8]. Four reference materials that are currently available for purchase (NIST materials: RM 8704, Buffalo River sediment and SRM 2710a, Montana I soil, and IAEA materials: SL-1, lake sediment and IAEA-385, Irish Sea sediment) have certified activities for

a variety of radionuclides but not for  $^{129}\text{I}$  have been identified. Previous work has shown that  $^{129}\text{I}$  is present in IAEA-385 [9]; however no literature values for  $^{129}\text{I}$  could be found for the other three materials. These materials have been analyzed at INL using two independent chemical separation and mass spectrometric analysis techniques. They involve an aqueous leaching preparation for analysis by accelerator mass spectrometry (AMS) and a combustion method of the starting material followed by thermal ionization mass spectrometry (TIMS) analysis. Both methods were validated via analyses of IAEA-375 for  $^{129}\text{I}$  and show good agreement with the recommended activity of  $1.7 \times 10^{-3} \text{ Bq kg}^{-1}$  for  $^{129}\text{I}$ ; ( $1.6 \times 10^{-3} \text{ Bq kg}^{-1}$  by AMS and  $1.8 \times 10^{-3} \text{ Bq kg}^{-1}$  by TIMS) with both sets of results within the 95% confidence interval of the recommended value [10].

## 2. Materials and methods

## 2.1. Preparation of samples for AMS

Details of the preparation of samples for AMS have been reported [11]. In brief, each standard was leached in a 5% tetramethylammonium

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hydroxide (TMAH) solution at approximately 90 °C for over ten hours. The size of aliquot leached was determined by the expected total iodine concentration in the standard. Approximately 1 – 2 g of each standard was leached in 30 – 75 mL of 5% TMAH. A reciprocating bath was used to aid the leach. An aliquot of each leachate was subsequently diluted and analyzed for  $^{127}\text{I}$  by inductively coupled plasma-mass spectrometry (ICP-MS). A second aliquot of the TMAH leachate was prepared for AMS analysis. Iodine carrier was added to this aliquot to adjust the  $^{129}/^{127}\text{I}$  ratio to approximately  $5 \times 10^{-11}$ . The carrier used was Inorganic Ventures 1000  $\mu\text{g I/mL}$  ion chromatography standard, with a  $^{129}/^{127}\text{I}$  ratio of approximately  $2 \times 10^{-13}$ . To ensure isotopic equilibration between the leachate and carrier, samples were taken through a series of redox steps. Aliquots of the resulting sodium bisulfite solution were added to an electrochemical cell where the iodide was deposited for two hours on a silver clad niobium wire working electrode [12]. The wire and silver iodide deposit were pressed into a titanium cathode for  $^{129}\text{I}$  analysis by AMS.

## 2.2. Preparation of samples for TIMS

Details of the preparation of samples for TIMS have been reported [10]. TIMS requires more stringent sample preparation than AMS in order to remove interferences. Unwanted contaminants can reduce analyte signal intensity due to competing ionization mechanisms, as well as introduce spectral interferences. The method is described briefly in this section. A key distinction between the AMS and TIMS sample preparation methods involves the use of  $^{125}\text{I}$  in the TIMS method. Approximately 1 g aliquots of each standard were spiked with  $^{125}\text{I}$ , loaded into combustion tubes and heated at 850 °C in an oxygen atmosphere for one hour. The  $^{125}\text{I}$  is used to determine the yield through the combustion chemistry as well as quantify the  $^{127}\text{I}$  and  $^{129}\text{I}$  in the TIMS measurement. It has been shown that the majority of the iodine is volatilized from soil samples at temperatures above 700 °C [13]. This initial step equilibrates the  $^{125}\text{I}$  with the sample as the combustion products transfer to a charcoal trap. The iodine was further purified chromatographically through a hydrated manganese dioxide bed with oxygen gas flow and at 600 °C to remove additional halides, such as, bromine and chlorine. The sample was collected back on a small volume of charcoal which was subsequently heated under nitrogen to drive the iodine into an ammonium hydroxide solution. Each sample was loaded onto a rhenium ribbon by in situ precipitation of silver iodide using silver nitrate for analysis by TIMS. Due to the amount of effort and time involved in the TIMS analyses the number of samples has been limited to two approximately 1 g aliquots for each of the reference materials.

## 2.3. Measurement by AMS

The instrument used to make the AMS measurements at INL is an NEC ICAMS which is a 0.5 MV compact accelerator mass spectrometer designed to analyze iodine in the +3 or the +2 charge state and is shown in Fig. 1. The conceptual design for the instrument was based on work performed at the Laboratory of Ion Beam Physics at ETH Zurich [14–16] which demonstrated the use of low energy compact accelerator mass spectrometers for detecting heavy ions such as  $^{129}\text{I}$ . Unlike the larger AMS instruments that typically select a higher charge state such as +5 for iodine and have relatively low transmission through the accelerator, the compact AMS instruments utilize the lower more probable charge states and can achieve transmissions of nearly 50% [16]. Helium, the preferred stripper gas for iodine, has the advantage of having less scattering losses through the accelerator while still providing adequate molecular interference suppression [17]. When using helium as a stripper gas and selecting the +2 charge state for analysis, the ion transmission in INL's instrument is ~40% for iodine. The instrument was installed at INL in 2015 and has been used for routine environmental iodine measurements since that time.

This instrument consists of a low energy injection magnet

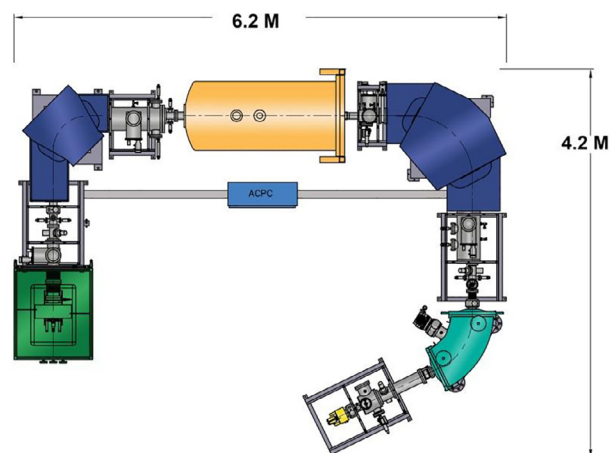


Fig. 1. NEC ICAMS (diagram courtesy of National Electrostatics Corp.).

( $r = 45.72\text{ cm}$ ) followed by the accelerator and then a high energy analyzing magnet ( $r = 75.0\text{ cm}$ ) followed by a 64° electrostatic analyzer (ESA). The ion source is a standard NEC 40 sample source of negative ions by cesium sputtering (SNICS). Samples are loaded as solid silver iodide that has been electrodeposited on 50  $\mu\text{m}$  diameter silver clad niobium wire using the same procedure reported by our lab for electrodeposition on silver wire [11] and pressed into titanium cathodes for targets in the NEC SNICS source. With the cesium oven temperature between 80° and 90 °C, the resulting high energy  $^{127}\text{I}^{+2}$  current ranges from 100 to 1000 nA. Low and high energy bouncing systems allow for iodine isotopic measurements of  $^{125}\text{I}$ ,  $^{127}\text{I}$  and  $^{129}\text{I}$ . The isotopic measurement is performed by rapidly switching ( $\sim 7\text{ Hz}$ ) the  $^{127}\text{I}^{+2}$  into an offset Faraday cup located after the high energy analyzing magnet and then switching the  $^{129}\text{I}^{+2}$  into a gas filled ionization detector installed after the ESA. INL does not currently use  $^{125}\text{I}$  in the AMS analysis of iodine instead uses ICP-MS for the  $^{127}\text{I}$  quantification in conjunction with carrier addition and the AMS measurement to quantify the  $^{129}\text{I}$  (Section 2.1). There are no technical reasons preventing INL from using a  $^{125}\text{I}$  spike to quantify the  $^{127}\text{I}$  and  $^{129}\text{I}$ . However, the high energy bouncing system is limited to  $\pm 20\text{ kV}$ . Therefore, in order to swing the  $^{129}\text{I}$  and the  $^{125}\text{I}$  into the gas filled ionization detector during a data collection cycle the terminal voltage would have to be reduced.

A series of quality control (QC) samples interleaved amongst the samples are loaded on the 40 position sample wheel to correct for background and normalize the data. The QC samples include matrix blanks, matrix blanks exposed to the sodium hydroxide electrodeposition, carrier blanks and INL-C normalization standards ( $^{129}/^{127}\text{I} = 5.62 (\pm 0.03) \times 10^{-11}$  ( $k = 1$ ) calibrated by our laboratory using NIST SRM 3230). Carrier blank ratio measurements are typically  $^{129}/^{127}\text{I} = 2 \times 10^{-13}$ . All reported data have been corrected for background and carrier blanks and the reported uncertainty is one standard deviation ( $k = 1$ ) of  $n$  measurements. Uncertainties of the normalization standard and ICP-MS data have been propagated.

## 2.4. Measurement by TIMS

TIMS measurements were performed on a custom designed triple sector instrument constructed around the VG Sector 54 platform using the VG source housing, two VG magnets and an ESA in a BBE-C configuration [18]. With a few modifications to the original design, this instrument has been used for iodine analysis at INL since 2003. The ion source uses a triple-filament arrangement [19] and analyzes negative ions to reduce hydride interferences. The instrument has an abundance sensitivity of  $\sim 10^{11}$ . However, unlike an AMS sputter source which can produce  $\mu\text{A}$  of  $^{127}\text{I}$  current, the TIMS source produces much less  $^{127}\text{I}$  current ( $\leq 10\text{ nA}$ ) which limits measurements of  $^{129}/^{127}\text{I}$  isotopic ratios to  $\sim 10^{-9}$ . Offset Faraday cups located after the first magnet allow for

**Table 1**  
Comparison of AMS and TIMS results, literature and IAEA-375 certificate.

IAEA 375	$^{129}\text{I}/^{127}\text{I}$	% uncertainty ( $k = 1$ )	$^{129}\text{I}$ Bq kg $^{-1}$	% uncertainty ( $k = 1$ )
AMS ( $n = 10$ )	$1.89 \times 10^{-7}$	2.77	$1.60 \times 10^{-3}$	2.52
TIMS ( $n = 2$ )	$1.61 \times 10^{-7}$	2.50	$1.80 \times 10^{-3}$	1.57
Jiang et al. [21]	n/a	n/a	$1.59 \times 10^{-3}$	2.52
Certificate	n/a	n/a	$1.70 \times 10^{-3}$	11.76

making simultaneous isotopic ratio measurements of  $^{129}\text{I}/^{127}\text{I}$  and then  $^{125}\text{I}/^{127}\text{I}$ . The minor isotopes are counted on a discrete dynode electron multiplier located after the ESA. Data is collected following a sequential collection protocol in which the signal intensity is systematically increased during the analysis. The slope of the linear regression applied to the plot of the  $^{129}\text{I}$  intensity vs the  $^{127}\text{I}$  intensity represents the  $^{129}\text{I}/^{127}\text{I}$  ratio and similarly for the  $^{125}\text{I}/^{127}\text{I}$  ratio. The  $^{125}\text{I}$  is used to determine the yield through the combustion chemistry as well as quantify the  $^{127}\text{I}$  and  $^{129}\text{I}$  in the TIMS measurement.

Quality control samples included with the TIMS samples are process blanks, NIST SRM 3230 ( $^{129}\text{I}/^{127}\text{I} = 0.982 (\pm 0.012) \times 10^{-8}$ ) standards and  $^{129}\text{I}/^{127}\text{I}$  blank solution from NIST SRM 3230 ( $^{129}\text{I}/^{127}\text{I} = 16 (\pm 5) \times 10^{-15}$  [20]). Cross calibration of the detectors is performed using a technique developed at INL which uses the minor oxygen isotopes of  $\text{ReO}_4^-$  to simultaneously calibrate the discrete dynode with the Faraday cups. Essentially, the major isotope at mass 251 is measured on a Faraday cup located after the first magnetic sector and the minor isotope at mass 255 is measured on the discrete dynode simultaneously (theoretical  $255/251 = 2.52 \times 10^{-5}$ ).

### 3. Results

#### 3.1. IAEA-375

IAEA-375 (soil contaminated with Chernobyl fallout) has a total iodine concentration of approximately  $1.4 \mu\text{g}_{(\text{I})} \text{g}^{-1}$  soil and a

**Table 2**  
Comparison of AMS and TIMS NIST RM 8704 results.

NIST RM 8704	$^{129}\text{I}/^{127}\text{I}$	% uncertainty ( $k = 1$ )	$^{129}\text{I}$ Bq kg $^{-1}$	% uncertainty ( $k = 1$ )
AMS ( $n = 5$ )	$1.48 \times 10^{-7}$	2.2	$1.41 \times 10^{-3}$	2.5
TIMS ( $n = 2$ )	$1.32 \times 10^{-7}$	0.5	$1.54 \times 10^{-3}$	1.8

recommended  $^{129}\text{I}$  value of  $1.7 (\pm 0.4) \times 10^{-3} \text{ Bq kg}^{-1}$ , and recently has been measured to be  $1.59 (\pm 0.08) \times 10^{-3} \text{ Bq kg}^{-1}$  [21] and  $1.6 (\pm 0.04) \times 10^{-3} \text{ Bq kg}^{-1}$  by AMS and  $1.8 (\pm 0.03) \times 10^{-3} \text{ Bq kg}^{-1}$  by TIMS see Table 1 [10]. In this previous study by INL IAEA-375 demonstrated the validity of the two methods reported in this paper. Since that time INL has used IAEA-375 as a control sample to assess the overall health of the process from the chemical purification through the AMS analysis. The  $^{129}\text{I}$  activity data obtained for 37 AMS analyses of IAEA-375 at INL is summarized in the control chart shown in Fig. 2 with the long term running average of  $1.56 (\pm 0.05) \times 10^{-3} \text{ Bq kg}^{-1}$ .

#### 3.2. Other reference materials

Four reference materials that are currently available for purchase (NIST materials: RM 8704, Buffalo River sediment and SRM 2710a, Montana I soil, and IAEA materials: SL-1, lake sediment and IAEA-385, Irish Sea sediment) have certified activities for a variety of radionuclides but not for  $^{129}\text{I}$ . Previous work has shown that  $^{129}\text{I}$  is present in IAEA-385 [9]; however no literature values for  $^{129}\text{I}$  could be found for the other materials. Previously, INL reported AMS and TIMS data for two additional reference materials NIST SRM 4354 and NIST SRM 4357. [10]

##### 3.2.1. NIST RM 8704

NIST RM 8704 (Buffalo River sediment) is very similar in composition to SRM 2704, having been collected at the same time and location. It has a total iodine concentration of approximately  $1.8 \pm 0.6 \mu\text{g}_{(\text{I})} \text{g}^{-1}$  soil [22]. Five, 1 g aliquots of NIST RM 8704 were

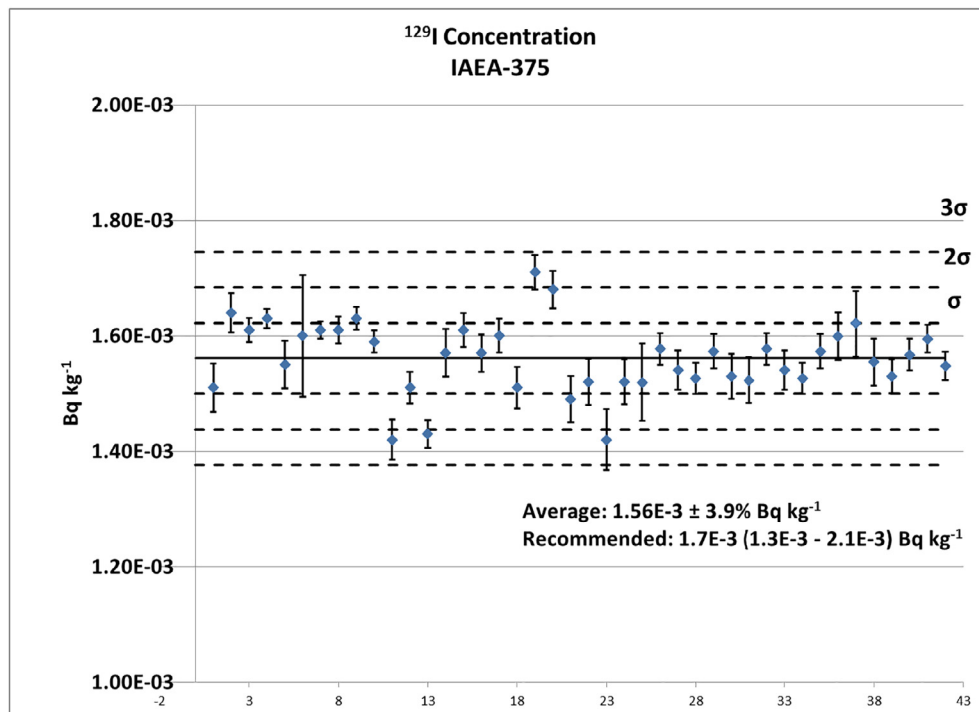


Fig. 2. IAEA-375 control chart.

**Table 3**  
Comparison of AMS and TIMS NIST SRM 2710a results.

NIST SRM 2710a	$^{129}\text{I}/^{127}\text{I}$	% uncertainty ( $k = 1$ )	$^{129}\text{I}$ Bq $\text{kg}^{-1}$	% uncertainty ( $k = 1$ )
AMS ( $n = 5$ )	$2.23 \times 10^{-9}$	27.3	$3.58 \times 10^{-5}$	1.6
TIMS ( $n = 2$ )	$1.34 \times 10^{-9}$	7.5	$3.50 \times 10^{-5}$	5.5

**Table 4**  
Comparison of AMS and TIMS SL-1 results.

IAEA SL-1	$^{129}\text{I}/^{127}\text{I}$	% uncertainty ( $k = 1$ )	$^{129}\text{I}$ Bq $\text{kg}^{-1}$	% uncertainty ( $k = 1$ )
AMS ( $n = 10$ )	$1.90 \times 10^{-9}$	8.5	$1.34 \times 10^{-4}$	15.1
TIMS ( $n = 2$ )	$1.68 \times 10^{-9}$	5.9	$1.15 \times 10^{-4}$	6.9

**Table 5**  
Comparison of AMS, TIMS results and IAEA 385 literature value [9].

IAEA 385	$^{129}\text{I}/^{127}\text{I}$	% uncertainty ( $k = 1$ )	$^{129}\text{I}$ Bq $\text{kg}^{-1}$	% uncertainty ( $k = 1$ )
AMS ( $n = 5$ )	$3.52 \times 10^{-8}$	2.4	$1.43 \times 10^{-2}$	1.3
TIMS ( $n = 2$ )	$2.87 \times 10^{-8}$	1.8	$1.58 \times 10^{-2}$	3.8
Daraoui [9]	$3.9 \times 10^{-8}$	5.1	$1.6 \times 10^{-2}$	4.1

leached in 30 mL of 5% TMAH for AMS analysis. Comparison of the AMS and TIMS results obtained is shown in Table 2.

### 3.2.2. NIST SRM 2710a

The NIST SRM 2710a, Montana I soil has a total iodine concentration of approximately  $3.1 \mu\text{g}_{(\text{I})} \text{g}^{-1}$  soil. Five, 0.5 g aliquots of NIST SRM

2710a were leached in approximately 30 mL of 5% TMAH for AMS analysis. The results from the AMS and TIMS analyses are shown in Table 3.

### 3.2.3. IAEA SL-1

The IAEA SL-1 (Lake sediment) has a reported total iodine concentration of approximately  $28 \mu\text{g}_{(\text{I})} \text{g}^{-1}$  soil on the IAEA reference sheet and a value reported in the literature of  $25.72 \pm 0.08 \mu\text{g}_{(\text{I})} \text{g}^{-1}$  [23]. Ten, 0.5 g aliquots of IAEA SL-1 were leached in 30 mL of 5% TMAH for AMS analysis. Table 4 shows the results from the AMS and TIMS analyses.

### 3.2.4. IAEA-385

The IAEA-385 (Irish Sea sediment) has a total iodine concentration of approximately  $62 \mu\text{g}_{(\text{I})} \text{g}^{-1}$  soil and  $^{129}\text{I}$  activity of approximately  $1.6 \times 10^{-2} \text{ Bq kg}^{-1}$  [9]. Five, 0.5 g aliquots of IAEA-385 were leached in 30 mL of 5% TMAH. Table 5 shows the results from the AMS and TIMS measurements.

## 4. Discussion

The two mass spectrometric techniques applied to the four environmental reference materials presented in this work show reasonable agreement with each other and when available agrees well with literature values. Certainly for some of the materials, such as NIST SRM 2710a and IAEA SL-1 the  $^{129}\text{I}/^{127}\text{I}$  ratio is approaching the limits of the TIMS measurement ( $10^{-9}$ ) which may explain some of the differences in the results. On the other hand, the ICP-MS measurement used in calculating the  $^{129}\text{I}/^{127}\text{I}$  ratio in the AMS method is prone to matrix effects that can influence the results. Also, homogeneity issues may be present based on the small aliquots of the reference materials used in this study.

These results combined with the results from our previous work [10], which includes NIST SRM 4354 and NIST SRM 4357, provides a very useful compilation of iodine concentrations for seven reference materials that cover over 3 orders of magnitude for  $^{129}\text{I}$  and nearly two

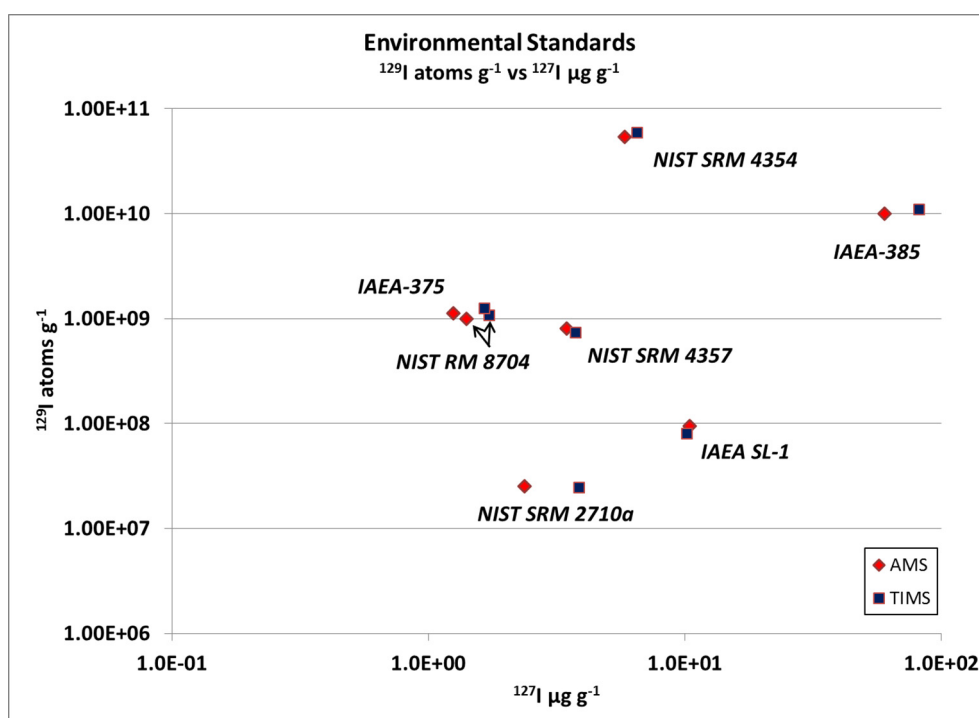


Fig. 3.  $^{129}\text{I}$  atoms  $\text{g}^{-1}$  vs.  $^{127}\text{I}$   $\mu\text{g g}^{-1}$  (error bars omitted for clarity).



orders of magnitude for  $^{127}\text{I}$  (Fig. 3). The plot ( $^{129}\text{I}$  atoms  $\text{g}^{-1}$  vs.  $^{127}\text{I}$   $\mu\text{g g}^{-1}$ ) shows that out of the environmental reference materials analyzed NIST RM 8704 is the closest reference material to IAEA-375. It also demonstrates the wide range of concentrations of iodine in these materials allowing a suitable standard to be selected to match the samples of interest.

## 5. Conclusions

The  $^{129}\text{I}$  activity and  $^{129}/^{127}\text{I}$  ratio have been quantified by two independent chemical and mass spectrometric measurement techniques, AMS and TIMS, in four different environmental reference materials: NIST RM 8704 (collected from the Buffalo River in the area of the Ohio Street Bridge, Buffalo, NY), NIST SRM 2910a (collected from the flood plain of the Silver Bow Creek approximately five miles west of Butte, Montana.), IAEA SL-1 (collected at the Sardis Reservoir; Panola County, Mississippi, USA) and IAEA 385 (collected at the Sardis Reservoir; Panola County, Mississippi, USA). In previous work both of these methods were validated using IAEA-375 which is the only soil/sediment material with a recommended  $^{129}\text{I}$  activity. Results from AMS ( $1.6 \times 10^{-3}$  Bq  $\text{kg}^{-1} \pm 2.52\%$ ) and TIMS ( $1.8 \times 10^{-3}$  Bq  $\text{kg}^{-1} \pm 1.57\%$ ) agreed very well with the IAEA-375 recommended value ( $1.7 \times 10^{-3}$  Bq  $\text{kg}^{-1}$ ) for  $^{129}\text{I}$  and both were within the 95% confidence interval on the certificate. The fact that IAEA-375 has not been available for purchase since 2010 demonstrates an immediate need in the AMS community for certified reference materials with known  $^{129}\text{I}$  concentrations. Although two independent methods were used for these measurements, it is only one source of data and by no means is enough to form any sort of consensus values. Therefore, the authors would invite other laboratories to undertake similar studies on these and other available materials to make  $^{129}\text{I}$  data more readily available in order to fill the void that the unavailability of IAEA-375 has created.

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