

## PRELIMINARY INVESTIGATION ON THE RAPID AND DIRECT AMS MEASUREMENT OF $^{129}\text{I}$ IN ENVIRONMENTAL SAMPLES WITHOUT CHEMICAL SEPARATION

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**ABSTRACT.** Accelerator mass spectrometry (AMS) is the most sensitive method for measuring  $^{129}\text{I}$  in environmental samples available today, with a detection limit of about  $10^{-15}$  for  $^{129}\text{I}/^{127}\text{I}$ . A drawback of the technique is the time-consuming chemical separation required to prepare AMS targets from raw samples. This step significantly limits applications requiring rapid analyses and large numbers of samples, for example, in monitoring studies associated with nuclear accidents. This work introduces a direct method for  $^{129}\text{I}$  measurements by AMS that does not require chemical separation. In this approach, stable iodine ( $^{127}\text{I}$ ) is added to a matrix of niobium (Nb) powder and mixed with dried raw sample. This mixture is pressed directly into a sputter target for AMS analysis. Two types of environmental samples have been tested in this work, seaweed and sediment. No anomalous behavior was noted in the  $\text{Cs}^+$  sputtering behavior of the targets prepared from these materials. The  $^{129}\text{I}/^{127}\text{I}$  ratios and  $^{129}\text{I}$  concentrations measured by this rapid method were found to be in agreement with reported values that used a conventional AMS method for the same material. Based on our findings, we expect that such rapid measurements can be applied to a wide variety of materials, in addition to seaweed and sediment, as long as the sputtering-induced adverse effects do not prevent the stable operation of the ion source. The method is especially useful for screening large numbers of samples before more precise analyses are made.

**KEYWORDS:** rapid and direct measurement, accelerator mass spectrometry,  $^{129}\text{I}$ , environmental samples, methodology.

### INTRODUCTION

Iodine-129 is a long-lived cosmogenic radionuclide, with a half-life of  $15.7 \times 10^6$  yr. In nature, it is produced by cosmic-ray interactions with Xe in the upper atmosphere, and by spontaneous fission of  $^{238}\text{U}$  and neutron-induced fission of  $^{235}\text{U}$  in the lithosphere (Edwards 1962; Eisenbud and Gesell 1997). The natural inventory has been estimated to be  $\sim 230$  kg, most of which resides in the deep oceans (Rao and Fehn 1999). However, the primary source of  $^{129}\text{I}$  in the Earth has now been significantly altered by human nuclear activities, and the natural concentration of  $^{129}\text{I}$  has been affected by continuing additions from a variety of anthropogenic sources (Santschi and Schwehr 2004). Civil and military nuclear activities and nuclear accidents have released more than 6000 kg of  $^{129}\text{I}$  since 1945 (Aldahan et al. 2007; Jabbar et al. 2013).

Due in part to its long half-life, production, and distribution,  $^{129}\text{I}$  has been recognized as a valuable radionuclide for environmental tracing studies and in assessing short- and long-term consequences of radioactive contamination in the environment (Yiou et al. 1994; Raisbeck et al. 1995; Raisbeck and Yiou 1999; Hou 2004; Santschi and Schwehr 2004; Fan et al. 2013). Among several analytical techniques capable of quantifying  $^{129}\text{I}$  in environmental samples, accelerator mass spectrometry (AMS) is now recognized as the method of choice for its superior sensitivity and small sample size requirements. However, the conventional AMS method for  $^{129}\text{I}$  determination requires time-consuming chemical separation of iodine. Typically, iodine is released from solid samples and trapped in aqueous solutions, then extracted and precipitated using wet chemistry procedures to produce AgI or AgI-AgCl (Hou et al. 2009, 2010). These procedures normally take more than a day, and the time required limits applications requiring rapid analyses, especially in emergency situations when the evaluation of a large number of

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samples is needed (e.g. following nuclear accidents). In these cases, it is desirable to use a simple and rapid preparation method to identify samples that can then be selected to slower but more precise analytical protocols. We describe below how a direct AMS measurement method for screening samples meets this need.

## Methodology

### **Standards, Blanks, and Target Materials**

The  $^{129}\text{I}$  standards used in the experiment were prepared by coprecipitating AgI-AgCl, with a Cl/I mass ratio of 20 and  $^{129}\text{I}/^{127}\text{I}$  atomic ratios of  $1.14 \pm 0.03 \times 10^{-10}$  (standard CST1) and  $9.95 \pm 0.32 \times 10^{-12}$  (standard CST2), using standardized  $^{129}\text{I}$  solutions and NaCl solutions. The  $^{129}\text{I}$  standard solutions were prepared by dilution of the NIST-SRM 4949c  $^{129}\text{I}$  standard (National Institute of Standards and Technology, USA) with a blank  $^{127}\text{I}$  solution, which is prepared by dissolution of crystalline iodine obtained from Woodward Iodine Corporation, USA, with an  $^{129}\text{I}/^{127}\text{I}$  ratio of  $<2 \times 10^{-13}$  (Hou et al. 2010; Zhang et al. 2013). All of the targets prepared in this study used 99.99% pure, 325-mesh niobium powder obtained from Alfa Aesar.

### **Samples**

Three certified reference materials, including sediment (IAEA-410) and seaweeds (IAEA-446, and NIST-SRM-4359) (Outola et al. 2006; Pham et al. 2006, 2014), were analyzed in this study. These materials are frequently used as quality control standard reference materials in  $^{129}\text{I}$  determinations. The  $^{127}\text{I}$  concentrations of the reference samples were analyzed by inductively coupled plasma mass spectrometry (ICP-MS) and their  $^{129}\text{I}$  activities were determined following standard procedures at the Xi'an AMS Laboratory (Zhang et al. 2011) for comparison with the direct method. Table 1 lists the concentration of  $^{129}\text{I}$  and  $^{127}\text{I}$ , and  $^{129}\text{I}/^{127}\text{I}$  atomic ratios in the samples.

### **AMS Target Preparation and Measurement**

The schematic diagram of the rapid AMS  $^{129}\text{I}$  detection procedure is presented in Figure 1. In the experiments, three types of targets have been tested: A: directly mixed sample powder with Nb, pressed into ordinary sample holders; B: Same as A, but the mixture was pressed into sample holders with enlarged holes; C: Same as B, but stable iodine was first added into the Nb before mixing with the dried sample powder. Two identical targets were prepared for each sample used in the tests of all three methods. The total mass of the mixture pressed into the targets and their iodine concentrations are listed in Table 1. Targets pressed with Nb powder only were used as machine blanks.

All targets were measured on the Xi'an 3.0MV AMS with the terminal voltage of 2.5MV and the charge state of +5, following the same steps as for the measurement of micro-iodine targets by the AgI-AgCl coprecipitation sample processing method, as described in Hou et al. (2010) and Liu et al. (2015). The ion source parameters and status are given in Table 2.

## RESULTS

Each target was measured sequentially, 10 min per cycle, for a total of six cycles. The machine blank level of  $^{129}\text{I}$  counts and digitized  $^{127}\text{I}$  ion charges collected were estimated using the average of corresponding Nb targets. The machine blank values were subtracted for each result to determine blank-corrected  $^{129}\text{I}/^{127}\text{I}$  ratios for each cycle. The final ratio of  $^{129}\text{I}/^{127}\text{I}$  was calculated as the average of multiple targets. The results are listed in Table 3.

Table 1 Information of sample and target preparation.

Target code	Raw sample information				Target preparation			Mass in the target	
	Sample material	Iodine concentration (µg/mg)	<sup>129</sup> I activity (mBq/g)	<sup>129</sup> I/ <sup>127</sup> I atom ratio	Mass ratio of Nb and sample (M <sub>Nb</sub> /M <sub>sample</sub> )	WWI added in Nb (µg/mg)	Iodine dilution coefficient	Total mass of the mixture pressed in holder (mg)	Iodine concentration (µg/mg)
S1-A-1/2	S1: Sediment	0.015	(1.29 ± 0.32) × 10 <sup>-5</sup>	(1.35 ± 0.34) × 10 <sup>-10</sup>	11.8	—	—	4.3/3.3	0.003
S1-B-1/2	IAEA-410	—	—	—	4.8	—	—	15.5/17.3	0.003
S1-C-1/2	—	—	—	—	8.4	7.4	4233.4	9.1/9.4	6.550
S2-A-1/2	S2: Seaweed	0.23	0.13 ± 0.02	(8.30 ± 1.3) × 10 <sup>-8</sup>	9.3	—	—	3.7/4.8	0.050
S2-B-1/2	IAEA-446	—	—	—	6.1	—	—	21.1/19.2	0.050
S2-C-1/2	—	—	—	—	6.8	7.4	212.8	8.6/7.4	6.423
S3-A-1/2	S3: Seaweed	1.098	0.0149 ± 0.002	(2.04 ± 0.27) × 10 <sup>-9</sup>	11.7	—	—	4.7/4.5	0.213
S3-B-1/2	NIST-SRM-4359	—	—	—	5.0	—	—	18.0/17.1	0.213
S3-C-1/2	—	—	—	—	8.7	7.4	60.1	10.6/7.6	6.688
Nb-1/2/3	Nb	—	—	—	—	—	—	18.1/19.1/16.9	—
Nb + WWI	Nb + WWI	—	—	<2 × 10 <sup>-13</sup>	—	7.4	—	15.3/20.9	7.400

Table 2 Parameters and status of cesium sputtering negative ion source (SO110).

Target voltage (kV)	6	Target current (mA)	0.1 ~ 0.15
Extraction voltage (kV)	29	Extraction current (mA)	0.08 ~ 0.1
Ionizer current (A)	17	Vacuum (mbar)	About 2 × 10 <sup>-7</sup>
Cesium reservoir temperature (°C)	75	Upper limit of vacuum during target changing (mbar)	About 2 × 10 <sup>-6</sup>

Table 3 Analytical results of the direct AMS measurements for <sup>129</sup>I.

Sample	<sup>129</sup> I/ <sup>127</sup> I ratio by conventional AMS method		Average <sup>129</sup> I/ <sup>127</sup> I ratio by rapid AMS method		<sup>129</sup> I activity directly measured by AMS (Method C) (mBq/g) (1σ)
	A	B	A	C*	
IAEA-410	(1.35 ± 0.34) × 10 <sup>-10</sup>	(0.86 ± 0.90) × 10 <sup>-10</sup>	(1.36 ± 0.84) × 10 <sup>-10</sup>	(2.12 ± 1.43) × 10 <sup>-10</sup>	(2.04 ± 1.38) × 10 <sup>-5</sup>
IAEA-446	(8.30 ± 1.30) × 10 <sup>-8</sup>	(10.3 ± 0.1) × 10 <sup>-8</sup>	(11.5 ± 0.2) × 10 <sup>-8</sup>	(10.2 ± 0.7) × 10 <sup>-8</sup>	(1.59 ± 0.11) × 10 <sup>-1</sup>
NIST-SRM-4359	(2.04 ± 0.27) × 10 <sup>-9</sup>	(2.56 ± 0.05) × 10 <sup>-9</sup>	(2.71 ± 0.03) × 10 <sup>-9</sup>	(2.02 ± 0.07) × 10 <sup>-9</sup>	(1.42 ± 0.05) × 10 <sup>-2</sup>

\*The <sup>129</sup>I/<sup>127</sup>I ratio for the original sample was calculated based on the measured <sup>129</sup>I/<sup>127</sup>I atomic ratio, the mass of the sample pressed in the holder and the amount of <sup>127</sup>I added in the Nb powder.

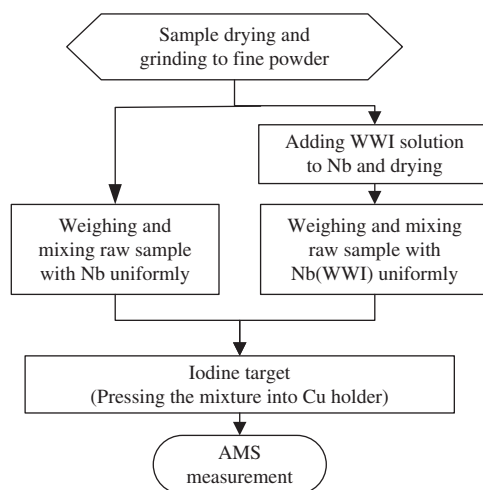


Figure 1 Schematic diagram of the analytical procedure for direct AMS measurement of  $^{129}\text{I}$ .

## DISCUSSION

The performance of this rapid method can be evaluated according to two criteria: (1) signal stability and (2) measurement reproducibility. A stable but increased intensity of the  $^{127}\text{I}^{5+}$  current with iodine concentration was observed for the suite of reference samples (Figure 2), and multiple targets of the same reference material produced consistent  $^{127}\text{I}$  (Figure 3) and  $^{129}\text{I}$  results (Figure 4). Therefore, these criteria are satisfied. The  $^{129}\text{I}/^{127}\text{I}$  ratios measured using the rapid method are also in general agreement with those of the conventional method including chemical pretreatment protocols, within their analytical uncertainties (Table 3).

### Sample Holder (Target) Design Improvement

In the first measurements (Method A), the dried sample material with a known iodine content was directly mixed with Nb powder, and the mixture (3.3–4.7 mg) was pressed in the standard sample holders with a bore diameter of 1.3 mm. This batch of targets including S1-A-1/2, S2-A-1/2, and S3-A-1/2 was run to test the stability of the ion source. Preliminary data (Figures 3, 4) for the  $^{129}\text{I}$  count rates and  $^{127}\text{I}$  currents showed promise, but the  $^{127}\text{I}$  signal was weak. This is partly attributed to the low total amount of iodine that was loaded in each target (see Table 1, IAEA-410: 0.003  $\mu\text{g}/\text{mg}$ ; IAEA-446: 0.050  $\mu\text{g}/\text{mg}$ ; NIST-SRM-4359: 0.213  $\mu\text{g}/\text{mg}$ ). We also reasoned that incomplete coverage of the slightly off-centered  $\text{Cs}^+$  density distribution over the target area could limit the number of  $^{127}\text{I}$  ions emitted from the ion source. As shown in Figure 5c, the  $\text{Cs}^+$  density distribution appears slightly off-centered, part of which appears to miss the sample area of 1.3 mm in diameter. Hence, the bore was enlarged to ensure the sample area to be fully exposed to the  $\text{Cs}^+$  beam (Figure 2b). The diameter of the bore in the new holder was increased from 1.3 to 2.1 mm, and the amount of sample mixture pressed into the holder was increased from about 15 to 21 mg. The increased sample material need is not an issue for such a rapid method, and this led to a 50% to 100% increase in iodine current. The details of the modification of the target holder are shown in Figure 5.

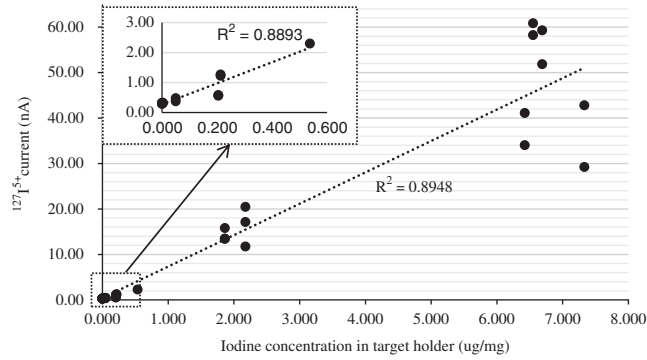


Figure 2 Correlation of the measured  $^{127}\text{I}^{5+}$  currents with iodine concentration in the targets.

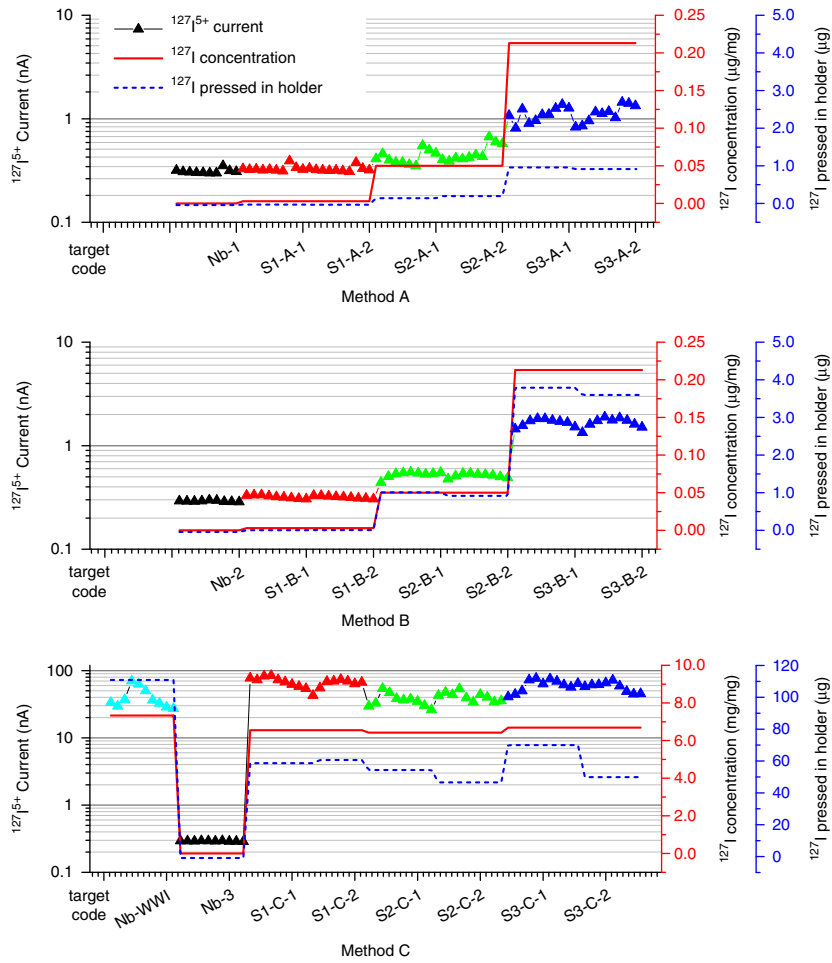


Figure 3 Comparison of the measured  $^{127}\text{I}^{5+}$  current and the  $^{127}\text{I}$  concentration/content in different targets of different methods.

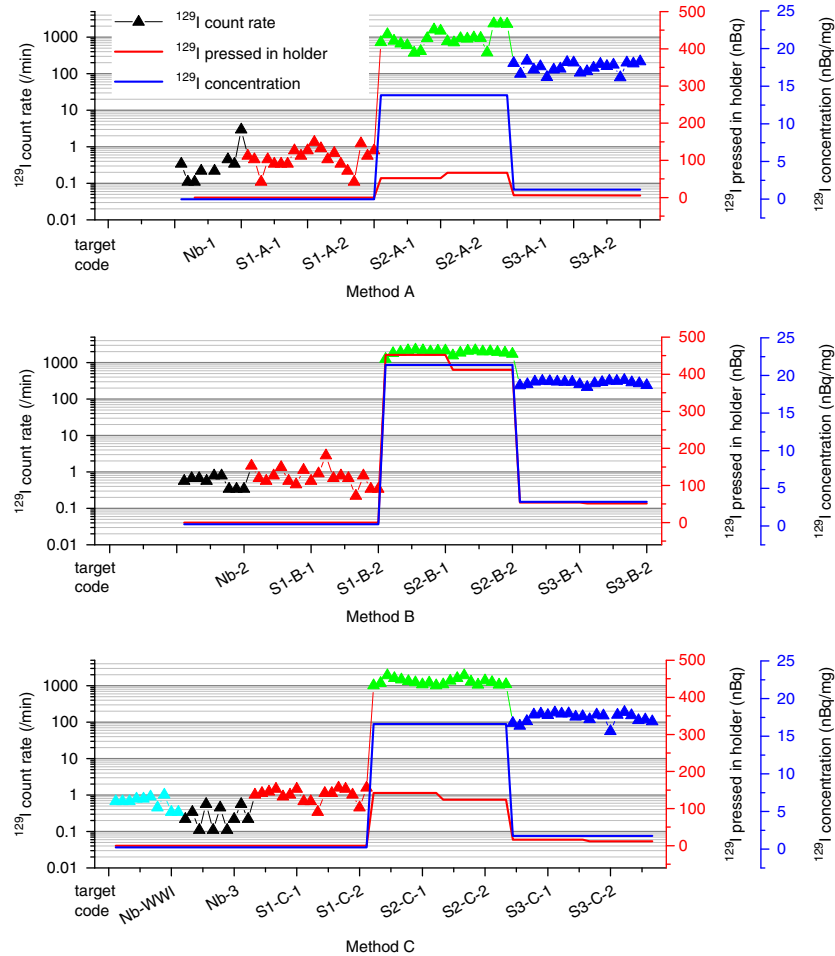


Figure 4 Comparison of the measured  $^{129}\text{I}$  count rate and the  $^{129}\text{I}$  concentration/content in different targets of different methods.

### Analytical Precision and Direct $^{129}\text{I}$ Activity Measurement by AMS

In Method A and B, the measured  $^{129}\text{I}/^{127}\text{I}$  ratios differ from known values notably because of generally low  $^{127}\text{I}$  currents. To improve the analytical accuracy of  $^{129}\text{I}$ , the measurement accuracy of  $^{127}\text{I}$  current has to be improved. To this end, a carrier addition to the target (Method C) was proposed. In this approach,  $^{127}\text{I}$  carrier solution was added to the Nb powder, the mixture was then homogenized, dried, and mixed with the reference samples for measurement. According to our previous work on AMS measurement of  $^{129}\text{I}$  in microgram-level iodine targets (Liu et al. 2015), when the iodine concentration reaches  $7\ \mu\text{g}/\text{mg}$  in the target, an  $^{127}\text{I}^{5+}$  current of  $30\sim 80\ \text{nA}$  can be measured at the high-energy end of the AMS; this  $^{127}\text{I}^{5+}$  current is sufficient to stabilize the  $^{127}\text{I}^{5+}$  beam position, and in turn to stabilize the terminal voltage by the slit stabilization function. In this work, the iodine concentration in the targets was increased by factors of 60, 213, and 4233 for NIST-SRM-4359, IAEA-446, and IAEA-410, respectively, with the addition of  $^{127}\text{I}$  carrier into the Nb powder. A precise  $^{129}\text{I}/^{127}\text{I}$  ratio was measured, in which the  $^{127}\text{I}$  from the raw sample contributed a small fraction ( $<1/60$ ). In this case, we can



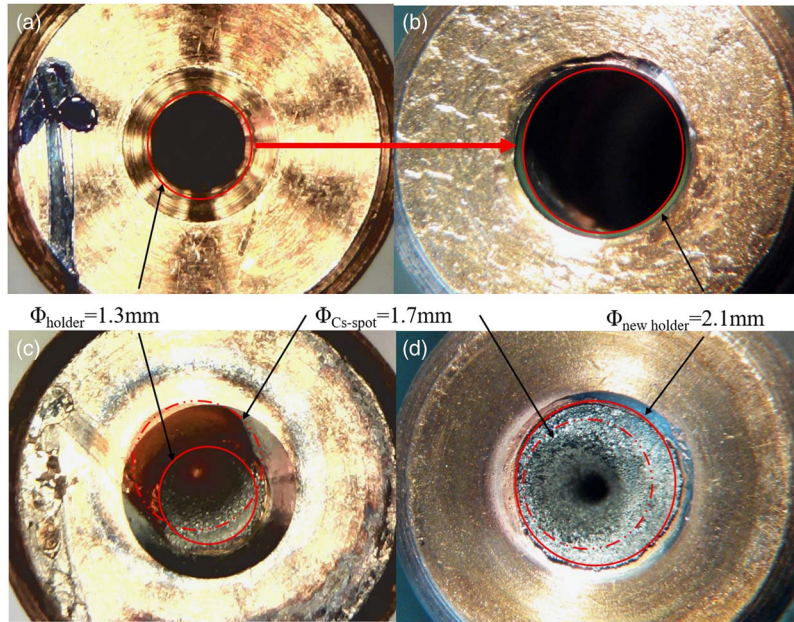


Figure 5 Sample holders (target): (a and b): original and improved target holders with different hole radii; (c and d): comparison of sputtered targets illustrating the efficient usage of sample material.

neglect the iodine content in the raw sample material and directly calculate the  $^{129}\text{I}$  activity concentration by the formula:

$$A_{129\text{I}} = \lambda_{129\text{I}} \times N_{129\text{I}}$$

$$\text{total } N_{129\text{I}} = \left( \frac{^{129}\text{I}}{^{127}\text{I}} \right)_{\text{measured}} \times (\text{total } N_{^{127}\text{I}})_{\text{in the target}}$$

$$\text{total } (^{127}\text{I})_{\text{in the target}} = \text{total } (^{127}\text{I})_{\text{from sample}} + \text{total } (^{127}\text{I})_{\text{from Nb powder}} \approx \text{total } (^{127}\text{I})_{\text{from Nb}}$$

$$\begin{aligned} (^{127}\text{I})_{\text{in the target}} \text{ corresponding to a unit mass sample} &\approx \frac{m_{\text{Nb}} * ^{127}\text{I}_{\text{concentration added in the Nb}}}{m_{\text{sample}}} \\ &= \frac{^{127}\text{I}_{\text{concentration added in the Nb}}}{m_{\text{sample}}/m_{\text{Nb}}} \end{aligned}$$

$$A_{129\text{I}} \text{ corresponding to a unit mass sample} = \lambda_{129\text{I}} \times \left( \frac{^{129}\text{I}}{^{127}\text{I}} \right)_{\text{measured}} \times \frac{R}{r} \times \frac{6.02 \times 10^{23}}{127}$$

in which  $\lambda_{129\text{I}}$  is the decay constant,  $R = \frac{^{127}\text{I}_{\text{concentration added in the Nb}}}{M_{\text{Pure Nb}}} = \frac{M_{\text{blank iodine}}}{M_{\text{Pure Nb}}}$  is the iodine concentration added to Nb, and  $r = \frac{m_{\text{sample}}}{m_{\text{Nb}}}$  is the mass ratio of sample and Nb (with blank iodine).

The  $^{129}\text{I}$  activity concentration was then calculated by the measured  $^{129}\text{I}/^{127}\text{I}$  ratio, combined with the mass of the raw sample, the mass of Nb powder added, and the concentration of iodine added to the Nb powder. The results (last column in Table 3) show that measured  $^{129}\text{I}$  concentrations are in excellent agreement with the reported values (fourth column in Table 1), within  $3\sigma$ .

#### Reliable Lower Detection Limit of $^{129}\text{I}$

The  $^{129}\text{I}$  concentration in NIST-SRM-4359 (target S3-C-1/2) is about 0.0015 mBq/g in the mixture loaded in the target, which resulted in a stable  $^{129}\text{I}$  count rate of about 100 counts/min with a statistical error of 5% for a 5-min measurement. Considering that the count rate of  $^{129}\text{I}$  for the machine's blank target (Nb) is about 1 count per minute (CPM), and we can set 10 times higher count rate (i.e. 10 CPM) for a reliable measurement of  $^{129}\text{I}$ , the lower detection limit can be set corresponding to the  $^{129}\text{I}$  concentration of 0.0015 mBq/g for the mixture. If the ratio of the raw sample to the Nb is maintained the same as the test samples, it can be concluded that the reliable detection limit of the method for accurate measurement of  $^{129}\text{I}$  activity is 0.0015 mBq/g in the raw sample materials. By this rapid method, assuming that all the conditions are kept the same as done in this experiment, if the count rate can get to 10 CPM or higher, we can directly report the  $^{129}\text{I}$  activity concentration; otherwise, we report that the  $^{129}\text{I}$  activity is lower than 0.0015 mBq/g in the raw sample. For the sample of IAEA-410 in this work, the  $^{129}\text{I}$  count rate is close to the machine background of Nb (about 1 CPM); we thus conclude that its  $^{129}\text{I}$  activity is lower than 0.0015 mBq/g, even though a value of  $(2.04 \pm 1.38) \times 10^{-5}$  (Table 3) is calculated after blank correction, which is close to the reference value.

#### Applicability

There were two types of samples tested in this work, seaweed and sediment. In this rapid measurement method, no particular anomaly was noted during  $\text{Cs}^+$  sputtering. The target integrity appeared quite normal and satisfied the need for direct AMS measurements. Based on this, it is expected that other environmental samples, such as soil, grass, and air particles, can be also analyzed using this method, as long they do not produce severe  $\text{Cs}^+$  sputter-induced adverse effects that prevent stable operation of the ion source. Because a small sample (1–3 mg) is used for analysis in this work, attention needs to be given to samples that might contain “hot” particles. Hot particles cause large variations in the analytical results, or errors in estimation of the average  $^{129}\text{I}$  level. Analyzing multiple sample subsets might be needed in this case.

It must be noted that because the stable iodine carrier added to the Nb powder and the  $^{129}\text{I}$  atoms bound by the raw samples are in different chemical states, isotope fractionation during  $\text{Cs}^+$  sputtering is possible. This is the fundamental reason why such a rapid method can only be used for screening or quick assessment purposes.

It is necessary to also note that such direct measurement was made using  $^{129}\text{I}^{+5}$  ions, for which the prime interfering molecular fragment is  $^{103}\text{Rh}^{+4}$  (Kilius et al. 1997). The abundance of Rh is extremely rare in Earth's crust, which is perhaps an important reason why direct  $^{129}\text{I}^{+5}$  counting could succeed without chemical purification of the target material. The direct detection of  $^{129}\text{I}$  using lower charge states may require additional caution in order to contend with more common molecular fragment interferences.

#### CONCLUSION

This work developed a direct AMS method for sensitive measurement of  $^{129}\text{I}^{+5}$  in environmental samples without chemical separation of iodine. The technique includes the addition of



$^{127}\text{I}$  carrier into a matrix powder (Nb), which is then mixed with dried raw sample powder. The mixture is then loaded directly into a modified target holder with an area of material exposure to fully catch the  $\text{Cs}^+$  spot. Seaweeds and sediment were tested as typical materials, and the results show that this direct AMS measurement method worked reasonably well for these two types of sample materials. The  $^{129}\text{I}/^{127}\text{I}$  ratios and  $^{129}\text{I}$  activities measured by the rapid method were found to be consistent with reported values within analytical uncertainties. It is therefore concluded that it is feasible to directly measure  $^{129}\text{I}/^{127}\text{I}$  ratios and  $^{129}\text{I}$  activities using AMS without chemical separation. It is expected that such a rapid measurement method can be also applied to other environmental solid materials, as long as the ion source can function stably. This would be especially useful in emergency situations, where rapid screening analyses of large numbers of samples are necessary to evaluate potential contamination.

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