Assessing the effect of laboratory environment on sample contamination for I-129 accelerator mass spectrometry

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Abstract

Environmental contaminations of $^{129}$I were continuously monitored in various sample preparation rooms for accelerator mass spectrometry at the University of Tsukuba. Monitoring of $^{129}$I was performed at the rooms that have several histories to treat samples, in order to compare with the results at the sample preparation rooms. Ambient levels of atmospheric $^{129}$I in each room were estimated from the measured concentrations in the alkali trap solutions. This article reports one year monitoring results for temporal changes of stable iodine ($^{127}$I) and $^{129}$I contamination rates into the alkali trap solutions. It was found that $^{129}$I were lower than approximately $10^4$ atoms cm$^{-2}$ day$^{-1}$ in the rooms where no-samples and samples with environmental background levels of $^{129}$I were only handled. Values from $10^4$ to $10^5$ atoms cm$^{-2}$ day$^{-1}$ were recorded in another room where environmental samples, such as the samples derived from nuclear-power-plant accident, were treated. Higher levels of $^{129}$I, ranging from $10^6$ to $10^7$ atoms cm$^{-2}$ day$^{-1}$, were recorded in rooms used for treating neutron-activated iodine. The experimental results show that the $^{129}$I level depended on the sample-preparation histories for $^{129}$I samples in respective rooms. It is possible to estimate $^{129}$I contamination risk from atmosphere to the samples by knowing $^{129}$I level in the preparation room.

**Keywords:** atmospheric $^{129}$I, sample preparation room, room contamination level, contamination risk, background level, Accelerator Mass Spectrometry (AMS)
Introduction

Accelerator mass spectrometry (AMS) is one of the high sensitivity analytical methods for measurement of environmental levels of long-lived radioisotopes such as $^{10}$Be, $^{14}$C, $^{26}$Al, $^{36}$Cl, $^{41}$Ca and $^{129}$I and estimated these isotopic ratios to the femto level. This analytical method is applied to the studies of geology and hydrology, $^{14}$C-dating, cosmic-ray exposure dating and tracing the migration of nuclear waste. As high sensitivity measurement is possible, it can in some cases lead to overestimates of concentrations mainly due to sample contamination, the memory effect and noise at measurement. A sample has a risk to be contaminated during its preparation from its environment and also from other samples. We focused in this work on the case of environmental contamination via a study of the contamination level in preparation rooms from atmosphere, as there is a risk that disturbed nuclides can be deposited on apparatus or samples from a contaminated sample-preparation environment.

Our research involved measuring $^{129}$I (half-life: $1.57 \times 10^7$ y) in various samples by AMS. This radionuclide’s presence in the atmosphere, hydrosphere and biosphere is attributed to spontaneous fission of uranium, historical nuclear weapon tests, and accidents in nuclear facilities, neutron activation analysis and interactions with cosmic rays. Iodine-129 is one of the most important radionuclides as a tracer in environment, the isotopic ratio of $^{129}$I/$^{127}$I have being reached to values of $10^{-10}$ to $10^{-4}$ in the environment from $10^{-12}$ in the pre-nuclear era. The chemistry of iodine is relatively complex; iodine takes a number of valence states, it is chemically reactive such as volatile and adsorptive, also forms various inorganic and organic compounds like $\Gamma^-$, $\text{IO}_3^-$ and $\text{CH}_3\text{I}$. Thus, the rooms used to prepare samples tend to become gradually contaminated through the chemical treatment of samples with high $^{129}$I concentrations. This contamination may lead to overestimates in subsequent AMS results. We found by experience that the higher $^{129}$I level samples had a tendency to make contaminate...
surroundings.

Therefore, in 2013, by a simple alkaline trap solution method, we began monitoring of $^{129}\text{I}$ in air in our rooms usually used for AMS sample preparations, to certify our measurements.\(^6\) The $^{129}\text{I}$ monitoring was also performed at rooms known as a high $^{129}\text{I}$ contaminated area as well as rooms unrelated with chemistry as background, in order to compare with the results at the sample preparation rooms. In this paper, we report one year monitoring results for the $^{127}\text{I}$ and $^{129}\text{I}$ contamination rates into the alkali trap solutions at various laboratory rooms in the University of Tsukuba, Japan. The monitoring helped a lot to know variation of $^{129}\text{I}$ contamination risk for AMS at each room and to certify our sample measurements.

Methods

Previous research\(^{1,6,17,22}\) has suggested that atmospheric iodine species can be reliably sampled and analyzed by air filtering. However, we did not mention atmospheric iodine species, and in the present work, stable ($^{127}\text{I}$) and $^{129}\text{I}$ naturally deposited from the atmosphere into alkaline trap solutions placed in the rooms was evaluated. Iodine-129 analyses were based on the literature.\(^{21,23,24}\) The details of the experimental procedures are described below.

Iodine collection using the trap solution

The solution for iodine trapping (referred to here as the “trap solution”) was prepared to collect iodine from the room air as follows. Organic alkaline solution TMAH (tetra methyl ammonium hydroxide, Tama Chemicals, Ultrapure Analytical Reagent, 25%) was diluted with Milli-Q water to 1.5–2% concentration. Approximately 1.0 or 1.5 L of the solution was transferred to a 2-L polyethylene bottle (outer diameter: 12.6 cm, height: 23.4 cm, mouth inner diameter: 7.5 cm). The amount of the trap solution before and after iodine collection were
obtained through weighing using an electric balance. Typical water volume evaporated during the iodine collection was approximately 20% for the initial volume.

From July 2013 to August 2014, the trap solutions in the open bottles were left in nine rooms described in next section, at the University of Tsukuba (36°06´N, 140°06´E), located on Tsukuba, Japan, for three weeks. This term was chosen because the duration should be long enough to allow various sample-preparation procedures to be conducted by various researchers. The trap solutions were placed on the floor in ICP-MS room and Passage in front of RI room and on the laboratory table in other seven rooms. The iodine collection was repeated up to eight times per room during one year.

The examined room

The details of the examined rooms were summarized in Table 1. C208, C304, C209, C210, and C108 are located in a building of University of Tsukuba Tandem Accelerator Complex. Non-RI room, RI room, ICP-MS room, and Passage in front of RI room are located in a building of the Center for Research in Isotopes and Environmental Dynamics. Especially, RI room, ICP-MS room, and Passage in front of RI room are located in a controlled area of radioactive isotopes (RI controlled area). The distance between the buildings is approximately 100 m. Although the rooms are distributed in two different buildings, users of the rooms are common within the AMS group of the University of Tsukuba.

We guessed that sample treatment histories in the rooms are important for contamination. Therefore, the rooms were categorized roughly into five types by their sample-treatment histories as shown in Table 1: (1) Personnel rooms, unrelated with chemistry. (herein, type-1 rooms); (2) laboratory rooms used to treat samples (except for soil) with low levels of $^{129}$I and to prepare AgI precipitates for AMS$^{21}$ (type-2 rooms); (3) laboratory rooms used to treat environmental samples from Fukushima after the Fukushima Daiichi Nuclear Power Plant
(FDNPP) accident\textsuperscript{19,20} (type-3 room); (4) laboratory rooms used to process environmental samples from Fukushima with higher radioactive levels than type-3 room (type-4 rooms); (5) laboratory rooms in the RI controlled area used to treat samples with high concentrations of $^{129}$I, including neutron-activated iodine, and other radioactive samples (type-5 rooms).

\textit{Determination of $^{127}$I collected in the trap solution by ICP–MS and estimation of contamination rate of $^{127}$I}

The $^{127}$I concentrations collected in the trap solutions were determined by inductively coupled plasma–mass spectrometry (ICP–MS) (Agilent 8800) system using an internal reference method with $^{133}$Cs for sensitivity correction. The $^{127}$I concentrations which were lower than "procedure blank" were treated as not-detected. Then, the total $^{127}$I weight in the original trap solution was obtained by multiplying the measured concentration by the whole weight of the original trap solution after the collection. Finally, the contamination rate of $^{127}$I in unit of ng cm$^{-2}$ day$^{-1}$ was estimated by dividing the total $^{127}$I weight in the trap solution by both area of bottle mouth (44 cm$^2$) and the collection term. The detection limit of $^{127}$I was 0.03 ppb (estimated from blank average plus 3$\sigma$) and the relative standard deviation, 1$\sigma$, were $\leq 20\%$, typical were $\leq 5\%$ for each samples in this study. This detection limit was corresponding to approximately 0.15 ng cm$^{-2}$ day$^{-1}$ of $^{127}$I contamination rates.

\textit{Chemical separation of iodine from the trap solutions for AMS}

After iodine was collected in the trap solutions, the bottle was shaken well to mix the solution. A portion of the solution (approximately 400 g) were transferred into a 500 mL beaker and added 2 mg of stable $^{127}$I carrier reagent (NaI solution, Orion ionplus\textsuperscript{8}, 0.1000 ± 0.0005 M) with an $^{129}$I/$^{127}$I ratio of $(1.7–1.8) \times 10^{-13}$,\textsuperscript{24,25} 5\% Na$_2$SO$_3$ solution to reduce iodate to iodide, completely. To ensure acidic conditions (pH $\leq$ 1), 16 M HNO$_3$ was added and then the solution
was transferred into a separatory funnel, iodide was oxidized to I\(_2\) by addition of 5% NaNO\(_2\) solution for purification by extraction to CCl\(_4\). Solvent extraction and back extraction of iodine were carried out twice per sample. Extracted iodide was precipitated as AgI, which was washed with NH\(_4\)OH and purified water (Milli-Q), dried at 110°C. Chemical preparation scheme is shown in Fig. S1 (Supporting Information).

The trap solutions collected from the type-4 and 5 rooms were handled similarly, ~100 g instead of 400 g solution was used, and were some parts of the above extraction method skipped in the handling of solutions collected after January 2014, because these solutions contained enough \(^{129}\)I to be measured with enough sensitivity, thus allowing only simple preparation. Consistent results were obtained from both methods, \(^{129}\)I contamination rate (atoms cm\(^{-2}\) day\(^{-1}\)) ; (3.0 ± 0.1) \times 10^5 and (3.5 ± 0.1) \times 10^5 at non-RI room, (6.2 ± 0.2) \times 10^6 and (6.4 ± 0.1) \times 10^6 at RI room, the latter were simple method, results of sampling period from 20, Jan. to 10, Feb. 2014. Chemical preparation scheme is shown in Fig. S2.

**Chemical separation of procedure blank and reagent blank**

The procedure blank was also obtained as AgI. Approximately 400g of Milli-Q water was processed with adding 2 or 4 mg of iodine, and treated similarly as the actual samples. A reagent blank of AgI was obtained by transferring 2.0 mg iodine into a 10 mL glass centrifuge tube, then adding AgNO\(_3\) solution. Chemical preparation scheme is also showed in Fig. S3.

**AMS for \(^{129}\)I and estimation of contamination rate of \(^{129}\)I**

The dried AgI was mixed with Nb metallic powder (325 mesh, chemical purify: 99.99%) in a ratio of 1:4 by weight and pressed into an Al cathode cone as an AMS target. Then, \(^{129}\)I/\(^{127}\)I ratio in the sample was determined by AMS at Micro Analysis Laboratory, Tandem accelerator, the University of Tokyo (MALT), Tokyo, Japan. A terminal voltage of 3.47 MV and the charge
state of 5+ were chosen for acceleration and detection, with an instrumental blank \(^{129}\text{I}/^{127}\text{I}\) ratio of \(<2 \times 10^{-14}\).\(^{26}\) The technique was certified using a Purdue-2 standard reference material (Z94-0596) provided by the Purdue Rare Isotope Measurement Laboratory (PRIME Lab) at Purdue University, IN, USA., an \(^{129}\text{I}/^{127}\text{I}\) ratio of \(6.54 \times 10^{-11}\) in the measurement,\(^{27}\) the value was revised in 2014.\(^{28}\) Overall uncertainty was less than 10%. The original \(^{129}\text{I}\) concentration (as atom g\(^{-1}\) day\(^{-1}\)) and \(^{129}\text{I}/^{127}\text{I}\) ratio in the trap solutions were determined \(^{127}\text{I}\) concentration obtained from ICP-MS and the \(^{129}\text{I}/^{127}\text{I}\) ratios obtained from AMS. These values were obtained by subtracting contribution from "procedure blank" which means \(^{129}\text{I}\) from the carrier, reagents and all experimental procedures. The \(^{129}\text{I}\) contamination rate in unit of atom cm\(^{-2}\) day\(^{-1}\) was multiplied the \(^{129}\text{I}\) concentration by the weight of the original trap solution after the three weeks iodine collection.

**Results and Discussion**

The contamination rate of \(^{127}\text{I}\)

The contamination rate of \(^{127}\text{I}\) were calculated by the values of \(^{127}\text{I}\) concentration in Table S1 (Supporting Information). Concentration of \(^{127}\text{I}\) was in the range of 0.0159–4.15 µg L\(^{-1}\) (Table S1), consistent with earlier studies.\(^{21}\) The level of \(^{127}\text{I}\) contamination rate was depended on each room and kept almost constant value, regardless of sampling period. Averages of the \(^{127}\text{I}\) contamination rates in unit of ng cm\(^{-2}\) day\(^{-1}\) were estimated to be 0.16 for C208, 0.23 for C304, 0.14 for C209, 0.36 for C210, 0.14 for C108, 0.21 for Non-RI room, 4.27 for RI room, 0.56 for ICP-MS, and 1.93 for Passage in front of RI room. The mode of the associated uncertainty value was approximately 10% within the range from 1% to 53%. The room air in C208 and C304 belong to type-1 unrelated with chemistry can represent outer environmental air. In the \(^{127}\text{I}\) contamination rate, the room airs in type-2, 3, and 4 rooms showed similar level to outer
environmental air. Earlier studies have suggested that iodine is present in the atmosphere at concentrations of 0.3–20 ng m\(^{-3}\).\(^\text{22,29}\) The amount of the \(^{127}\text{I}\) collected in the trap solution was obviously smaller than amount of \(^{127}\text{I}\) in whole room air as same as outer environmental air. The \(^{127}\text{I}\) contamination rates at type-5 were significantly higher than the others. In these rooms, more amount of iodine reagents was used for chemical experiments than another-type rooms. The high \(^{127}\text{I}\) contamination rates attributed to the use of the reagents.

The contamination rate of \(^{129}\text{I}\)

The measured \(^{129}\text{I}\) contamination rates to the trap solutions at each room are plotted in Fig. 1. The \(^{129}/^{127}\text{I}\) ratios of reagent blank were \((2.0 \pm 0.6) \times 10^{-13}\), error means \(1\sigma \ (n = 6)\), and procedure blanks were \((2.0–5.6) \times 10^{-13}\). Our result of \(^{129}/^{127}\text{I}\) ratio \((2.0 \pm 0.6) \times 10^{-13}\) agrees well with the reference value\(^\text{24,25}\) in the iodine reagent (Orion ionplus\(^\text{®}\)) found by AMS at MALT has been reported as being around \(1.7 \times 10^{-13}\). The contamination rate of \(^{129}\text{I}\) were calculated by the values of \(^{129}\text{I}\) concentration in Table S1. The associated uncertainty values of \(^{129}\text{I}\) contamination rates accompanied with analytical procedures were within the range from 2\% to 60\%. The \(^{129}\text{I}\) contamination rates to the trap solutions were quite different among the rooms. In 2014, the \(^{129}\text{I}\) contamination rates in the type-1 rooms were consistent with environmental level, obtained \((0.67-1.2) \times 10^{4}\) atoms cm\(^{-2}\) day\(^{-1}\), that the \(^{129}\text{I}\) level of atmospheric fallout samples were \((0.92-31.3) \times 10^{4}\) atoms cm\(^{-2}\) day\(^{-1}\); calculated from \((0.39-13.3) \times 10^{-2}\) mBq m\(^{-2}\) month\(^{-1}\) recorded from 1986 to 2005,\(^\text{30}\) \((0.65-24) \times 10^{4}\) atoms cm\(^{-2}\) day\(^{-1}\) from 2009 to 2010 and \((1.0-12) \times 10^{4}\) atoms cm\(^{-2}\) day\(^{-1}\) during 2012,\(^\text{21}\) in Tsukuba, Japan. Although the \(^{129}\text{I}\) contamination rates at C209, \((0.41-3.2) \times 10^{4}\) atoms cm\(^{-2}\) day\(^{-1}\), were comparable with those at type-1 rooms as environmental circumstance, the \(^{129}\text{I}\) contamination rates at C210, \((1.0-26) \times 10^{4}\) atoms cm\(^{-2}\) day\(^{-1}\), and C108, \((0.56-16) \times 10^{4}\) atoms cm\(^{-2}\) day\(^{-1}\), were a little higher than those at type-1 rooms. The \(^{129}\text{I}\) contamination rates at Non-RI room, \((9.9-82) \times 10^{4}\) atoms
The measured $^{129}$I contamination rates pertain to the rooms’ histories. As we expected, treated samples with higher $^{129}$I concentrations tended to be induced higher $^{129}$I contamination.

Figure 1 also implies that in November and December of 2013, all the rooms were strongly contaminated by $^{129}$I. This was clearly due to the treating of high-concentration $^{129}$I solutions ($^{129}$I $\approx 10^{16}$ atoms g$^{-1}$, $^{129}$I/$^{127}$I $\geq 10^{-4}$) in the RI controlled area during these months.

The high-concentration $^{129}$I solutions were picked out from a shielded storage chamber and diluted a part of them to some concentrations at RI room. Then, the diluted solutions were transferred to ICP-MS room, performed ICP-MS for $^{127}$I and $^{129}$I, and stored there for approximately a month. Needless to say, we had kept the rules for behaviors in the RI controlled area (e.g., used a protective glove and washed our hands). However, the associated contamination was observed simultaneously at all the rooms, located outside of the RI controlled area in even different building. It was attributed to $^{129}$I, its active and volatile behavior, being diffused by air and human movement. Fortunately, as shown in the variations of $^{129}$I contamination rates at C209, C210, C108, Non-RI room, and RI room in Fig. 1, the level of $^{129}$I contamination decreased with time and returned to the level before the contamination was occurred. It could have temporary effects across the laboratory.

The variations of $^{129}$I/$^{127}$I ratios in the original trap solution at each room were plotted in Fig. 2. The variations of $^{129}$I/$^{127}$I ratios followed the $^{129}$I contamination rates shown in Fig. 1. After the strong contamination by $^{129}$I in November and December of 2013, the $^{129}$I/$^{127}$I ratios were also increased at that period and decreased with time. Then, the $^{129}$I/$^{127}$I ratio returned to be the ratio before the increase and became steady. The plot for the ICP-MS room appears different
from those for the other two rooms of type-5 with similar histories. This room was extended recently (in 2013) inside a building. Therefore, $^{129}$I contamination was strong, but $^{127}$I contamination was not so progresses and the $^{129}$I/$^{127}$I ratio in ICP-MS room could be enhanced higher than other rooms at same building. With the exception of the values obtained during 18 Nov. to 9 Dec. 2013, the $^{129}$I/$^{127}$I ratio at rooms of history types-1 to 3 ranged around $2 \times 10^{-8}$, which were also consistent with the $^{129}$I/$^{127}$I ratios of environmental level, from atmospheric fallout samples: before the FDNPP accident, $(0.66-23.4) \times 10^{-8}$ recorded from 1986 to 2005$^{30}$ and $(0.3-3.8) \times 10^{-8}$ recorded from 2009 to 2010,$^{21}$ after the FDNPP accident, $(1.1-5.8) \times 10^{-8}$ recorded 2012$^{21}$ in Tsukuba, Japan.

**Contribution from contamination to measurement of $^{129}$I/$^{127}$I ratio**

Ambient $^{129}$I is expected to contaminate any actual samples treated in that environment. The $^{129}$I contamination risk from atmosphere was defined as $^{129}$I/$^{127}$I ratio obtained by AMS, according to the following equation;

$$
R = \frac{^{129}\text{I precipitated from atmosphere (atoms)}}{^{127}\text{I carrier (atoms)}} = \frac{(C \times A \times T)}{(W \times N_A/127)}
$$

where $R$ is contribution of the $^{129}$I contamination from room air to $^{129}$I/$^{127}$I ratio, $C$ is the $^{129}$I contamination rate from room air (atoms cm$^{-2}$ day$^{-1}$), $A$ is the open area of apparatus (cm$^2$, e. g. beaker or centrifuge tube), $T$ is the working time (day), $W$ is the carrier weight (g) and $N_A$ is Avogadro constant. Here, within the results of the contamination rate of $^{127}$I, the $^{127}$I weight from atmosphere (ng range) is much less than the weight of carrier iodine (mg range). Therefore, the amount of $^{127}$I from atmosphere is negligible for $^{129}$I determination by AMS.

Figure 3 indicates variations of $^{129}$I/$^{127}$I ratio in procedure blank estimated from $^{129}$I contribution (R) of the various $^{129}$I contamination rate by Eq. (1). Here, we plugged in 10 cm$^2$ (e. g. beaker or centrifuge tube), 2 mg (general carrier weights for AMS), and 1hour, 0.5 day, 1day
and three weeks for parameters $A$, $W$, and $T$, respectively. The value from carrier reagent (reagent blank), $1.7 \times 10^{-13}$ is shown by a broken line in Fig. 3. In the case of C108 indicated $^{129}$I contamination rate among rooms where we usually used for AMS sample preparation, the contribution to $^{129}/^{127}$I ratio ($R$) was estimated to be $1.3 \times 10^{-14}$ by Eq. (1) using $^{129}$I contamination rate ($C$); $2.5 \times 10^4$ atoms cm$^{-2}$ day$^{-1}$ (average in the steady period from Fig. 1) and the working time ($T$); 0.5 day. Therefore, relative $^{129}$I contribution to the $^{129}/^{127}$I atoms ratio of procedure blank was 0.08, and estimated value of $^{129}/^{127}$I ratio in procedure blank is approximately $2 \times 10^{-13}$ for C108. This value, 0.08, fell within the error range of AMS. Actually, our results of procedure blanks from July 2013 to August 2014 were consistent with this contamination estimation. These results lead to conclusion that the level of $^{129}$I inventory in C209, C210, and C108 has no serious influence on our sample preparation. In the case of RI room, $R$ was estimated to be $2.2 \times 10^{-12}$ by Eq. (1) using $C$; $4.1 \times 10^6$ atoms cm$^{-2}$ day$^{-1}$ (average in steady period) and $T$; 0.5 day. Contribution of $^{129}$I estimated as contamination risk becomes ten times higher than reagent blank. Therefore, it was found that a measured value can be overestimated in case of sample preparation at RI room. At the high contaminated period in November and December of 2013, by the same calculation, relative $^{129}$I contribution to the $^{129}/^{127}$I atoms ratio of procedure blank was estimated to be 0.4 for C108. Although this factor is considerably high for accurate measurement, we had not prepared any sample during the period fortunately. The excess measured above the $(2.0 \pm 0.6) \times 10^{-13}$ background of the carrier reagent indicates atmospheric contamination of $^{129}$I in room or possibly some other reason such as too long time for preparation, cross contamination and the other. However, at least, we can estimate $^{129}$I contamination risk roughly from atmosphere in this method.

Conclusions
By the simple method using alkaline trap solutions, over one year, we had monitored $^{127}$I and $^{129}$I contamination rates from the airs not only at the three working rooms for AMS sample preparation but also two rooms unrelated with chemistry and four rooms expected as high $^{129}$I contaminated areas. It was found that the $^{127}$I contamination with ng-order weight from atmosphere was constant at each room regardless of sampling period and negligible for AMS compared with mg-order weight of carrier. In contrast, the $^{129}$I contamination rates strongly depended on the sample-preparation history. This fact implied that treated samples with higher $^{129}$I concentrations tended to be induced higher $^{129}$I contamination. Using the measured $^{129}$I contamination rate, in the rooms used usually for our AMS sample preparation, we certified that the contribution of $^{129}$I contamination from atmosphere for AMS was within error range of AMS. Furthermore, a simultaneous high $^{129}$I contamination event at seven rooms was observed clearly in variations of the $^{129}$I contamination rates. Therefore, this method has a high sensitivity for variation of the $^{129}$I contamination rate. As a conclusion of this study, this monitoring method helped greatly to know variation of $^{129}$I contamination risk for AMS and to validate our sample measurements. We intend to continue the monitoring of $^{129}$I in the atmosphere at AMS sample preparation rooms by this method.

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Supporting Information

There are sampling date, weight of trap solutions, $^{129}$I and $^{127}$I concentration in trap solutions, $^{129}$I/$^{127}$I ratio and $^{129}$I and $^{127}$I contamination rate for each samples in Table S1. Chemical schemes are in Fig. S1 – Fig. S3.

This material is available free of charge on the Web at http://www.jsac.or.jp/analsci/.
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### Table 1

Details of experimental rooms measured $^{129}$I contamination rates.

| Building name | Room name | Capacity (m$^3$) | Type | Purpose of room use | Kind of sample | Concentration of $^{129}$I (atoms g$^{-1}$) | Ratio of $^{129}$I/$^{127}$I | Duration of use |
|---------------|-----------|-----------------|------|---------------------|----------------|--------------------------------|-----------------|----------------|
|               | C208      | 75.2            | 1    | Desk work           |                | —                             | —               | —              |
|               | C304      | 150             | 1    | Desk work           |                | —                             | —               | —              |
| A             | C209      | 155             | 2    | Sample preparation  | Environmental for $^{36}$Cl and final preparation of AgI and AgCl | $10^7$ to $10^{10}$ | $10^{-11}$ to $10^{-8}$ | >20 years |
|               | C210      | 110             | 2    | Sample preparation  | Environmental water | $10^7$ to $10^{10}$ | $10^{-11}$ to $10^{-5}$ | >20 years |
|               | C108      | 196             | 3    | Sample preparation  | Environmental and soil for $^{129}$I, environmental samples for $^{14}$C | $10^7$ to $10^{10}$ | $10^{-11}$ to $10^{-5}$ | 4 years |
| B             | Non-RI room | 114            | 4    | Sample preparation  | Soil            | $10^9$ to $10^{10}$ | $10^{-11}$ to $10^{-5}$ | >20 years |
|               | RI room  | 164             | 5    | Sample preparation  | Soil etc.       | $<10^8$ | $10^{-4}$ or more | >20 years |
|               | B (Radiation controlled area) | ICP-MS room | 31.4 | measurement | Liquid for ICP-MS | $<10^{13}$ | $10^{-4}$ or more | 1 year |

History time 1 to 3 had low-level $^{129}$I use and are located in a building at (A) University of Tsukuba Tandem Accelerator Complex. History type 4 and 5 had high-level $^{129}$I use and are located in a building at (B) the Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba.
Figure captions

**Fig. 1** Variation of the measured $^{129}$I contamination rates to the trap solutions at each room. The symbol was plotted at a middle time of the sampling period on the vertical axis. Gray zones indicate background level from precipitation at Tsukuba. The numbers in parentheses mean room type, respectively.

**Fig. 2** Variation of measured $^{129}I/^{127}I$ ratios in the trap solutions at each room. The symbol was plotted at a middle time of the sampling period on the vertical axis. Gray zones indicate background level from precipitation at Tsukuba. The numbers in parentheses mean room type, respectively.

**Fig. 3** Variations of $^{129}I/^{127}I$ ratios in the procedure blank (Y-axis) adjusted contribution of $^{129}$I contamination (R) from room air estimated by Eq. (1), as a function of a variety of $^{129}$I contamination rates (C, X-axis) at different working time (T). The ratio of reagent blank is shown by a broken line, as ‘Blank level’, $1.7 \times 10^{-13}$. Here, $W = 2$mg and $A = 10 \text{ cm}^2$. 
Figure 1
Figure 2

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Figure 3

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Assessment of I-129 level in sample preparation rooms.

Iodine trapping

Accelerator Mass Spectrometry

I-129 Assessment

Tandem Accelerator

I-129 in atmosphere