Notes

Assessing the Effect of Laboratory Environment on Sample Contamination for I-129 Accelerator Mass Spectrometry

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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 in the rooms used for the treatment of samples in the past, in order to compare with the results obtained in 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 the results of one year of monitoring the temporal changes of stable iodine (^{127}I) and ^{129}I contamination rates in the alkali trap solutions. It was found that ^{129}I contamination rates were lower than approximately 10^4 atoms cm^{-2} day^{-1} in the rooms where ether no samples or only samples with environmental background levels of ^{129}I were 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 accidents, 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 ^{129}I sample-preparation histories for the respective rooms. It is possible to estimate the ^{129}I contamination risk from the atmosphere to the samples by knowing the ^{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)

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Introduction

Accelerator mass spectrometry (AMS) is one of the highly sensitivity analytical methods for measuring environmental levels of long-lived radioisotopes, such as ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca and ^{129}I, and estimating 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.[1,2] As a highly sensitive measurement is made possible by this method, it can in some cases lead to overestimates of concentrations, mainly due to sample contamination, the memory effect, and noise at measurement.[3-5] A sample has a risk of contamination during its preparation from its environment and also from other samples. In this work, focused on environmental contamination *via* a study of the contamination level in preparation rooms due to contamination of the 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: 1.57×10^7 y) in various samples by AMS.[6] This radionuclide's presence in the atmosphere, hydrosphere and biosphere is attributed to spontaneous fission of uranium, nuclear weapon tests, 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 the environment;[7-9] the isotopic ratio of ^{129}I/^{127}I has reached values of 10^{-10} to 10^{-4} in the environment from 10^{-12} in the pre-nuclear era.[10-12] The chemistry of iodine is relatively complex. Iodine takes a number of valence states; it is chemically reactive, being volatile and adsorptive, but also forms various inorganic and organic compounds like I^-, IO_3^- and CH_3I.[12-17] Thus, the rooms used to prepare samples tend to become gradually contaminated through the chemical treatment of samples with high ^{129}I concentrations.[18] 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 contaminate the surroundings.

Therefore, in 2013, by a simple alkaline trap solution method, we began monitoring of ^{129}I in the air in our rooms usually used for AMS sample preparations, to certify our measurements.[19-21] The ^{129}I monitoring was also performed in rooms believed to have high ^{129}I contamination, as well as rooms unrelated to chemistry, in order to compare with the results for the sample preparation rooms. In this paper, we report the results of one year of monitoring for the ^{127}I and ^{129}I contamination rates in the alkali trap solutions placed in various laboratory rooms at the University of Tsukuba, Japan. The monitoring greatly helped to discern the variation of ^{129}I contamination risk for AMS in each room and to certify our sample measurements.

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Table 1 Details of experimental rooms measured for ^{129}I contamination rates

Research area			History of the room use		Treated sample			
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
A	C208	75.2	1	Desk work	—	—	—	—
	C304	150	1	Desk work	—	—	—	—
	C209	155	2	Sample preparation	Environmental water for ^{36}Cl and final preparation of AgI and AgCl	10^7 to 10^{10}	10^{-11} to 10^{-5}	>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 water 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
B (Radiation controlled area)	RI room	164	5	Sample preparation	Soil *etc.*	$<10^{16}$	10^{-4} or more	>20 years
	ICP-MS room	31.4	5	Measurement	Liquid for ICP-MS	$<10^{13}$	10^{-4} or more	1 year
	Passage in front of RI room	—	5	Access to RI room	—	—	—	—

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.

Methods

Previous research[16,17,22] has suggested that atmospheric iodine species can be reliably sampled and analyzed by air filtering. However, we did not monitor atmospheric iodine species. In the present work, stable (^{127}I) and ^{129}I naturally deposited from the atmosphere into alkaline trap solutions placed in the rooms were evaluated. Iodine-129 analyses were based on earlier 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 amounts of the trap solution before and after iodine collection were obtained through weighing using an electric balance. The average water volume evaporated during the iodine collection was approximately 20% of the initial volume.
From July 2013 to August 2014, the trap solutions in open bottles were left in nine rooms described in the next section, at the University of Tsukuba (36°06′N, 140°06′E) located in Tsukuba, Japan, for three weeks. This length of time 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 an ICP-MS room, in the passage in front of an RI room, and on the laboratory table in seven other rooms. The collection of iodine was conducted up to eight times per room during the course of one year.

The examined rooms
The details of the examined rooms are summarized in Table 1. C208, C304, C209, C210, and C108 are located in a building of the 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. To be noted is that the 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, all of the rooms are used by various personnel within the AMS group of the University of Tsukuba.
We surmised that the sample treatment histories of the rooms are an important factor of 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, type1 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] (type2 rooms); (3) laboratory rooms used to treat environmental samples from Fukushima after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident[19,20] (type3 room); (4) laboratory rooms used to process environmental samples from Fukushima with higher radioactive levels than those processed in the type3 room (type4 room); (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 (type5 rooms).

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 an 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 that were lower than the "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 units of ng cm^{-2} day^{-1} was estimated by dividing the total ^{127}I weight in the trap solution by both the area of the bottle mouth (44 cm^2) and the collection period. The detection limit for ^{127}I was 0.03 ppb (estimated from blank average plus 3σ) and the relative standard deviation, 1σ, were $\leq 20\%$ in this study. This detection limit corresponded to a ^{127}I contamination rate of approximately 0.15 ng cm^{-2} day^{-1}.

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) was transferred into a 500-mL beaker and to this was added 2 mg of stable ^{127}I carrier reagent (NaI solution, Orion ionplus®, 0.1000 ± 0.0005 M) with an $^{129}I/^{127}I$ ratio of $(1.7 - 1.8)\times 10^{-13}$,[24,25] and 5% Na_2SO_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, and 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_4OH and purified water (Milli-Q), then dried at 110°C. The chemical preparation scheme is shown in Fig. S1 (Supporting Information).

The trap solutions collected from the type4 and type5 rooms were handled similarly, however, ~100 g instead of 400 g solution was used, and some parts of the above extraction method were skipped in the handling of solutions collected after January 2014, because these solutions contained enough ^{129}I to be measured with enough sensitivity. Consistent results were obtained from both methods; ^{129}I contamination rate (atoms cm^{-2} day^{-1}); $(3.0 \pm 0.1)\times 10^5$ and $(3.5 \pm 0.1)\times 10^5$ in non-RI room, $(6.2 \pm 0.2)\times 10^6$ and $(6.4 \pm 0.1)\times 10^6$ in RI room, with the latter results obtained from the simplified method. The results are for the sampling period from 20, Jan. to 10, Feb. 2014. The chemical preparation scheme is shown in Fig. S2 (Supporting Information).

Chemical separation of procedure blank and reagent blank

The procedure blank was also obtained as AgI. Approximately 400 g of Milli-Q water was processed with the addition of 2 or 4 mg of iodine, and treated similarly as the actual samples. A reagent blank of AgI was obtained by transferring 2 mg iodine into a 10-mL glass centrifuge tube, then adding $AgNO_3$ solution. The chemical preparation scheme is shown in Fig. S3 (Supporting Information).

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, the $^{129}I/^{127}I$ ratio in the sample was determined by AMS at the 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 $^{129}I/^{127}I$ 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, with an $^{129}I/^{127}I$ ratio of 6.54×10^{-11} in the measurement[27] (the value was revised in 2014).[28] Overall

uncertainty was less than 10%. The original ^{129}I concentration (as atoms g^{-1} day^{-1}) and $^{129}I/^{127}I$ ratio in the trap solutions were determined, with the ^{127}I concentration obtained from ICP-MS and the $^{129}I/^{127}I$ ratios obtained from AMS. These values were obtained by subtracting the contribution from the "procedure blank", which means ^{129}I from the carrier, reagents and all experimental procedures. The ^{129}I contamination rate in units of atoms cm^{-2} day^{-1} was multiplied by the ^{129}I concentration by the weight of the original trap solution after the three weeks of iodine collection, because water evaporates.

Results and Discussion

The contamination rate of ^{127}I

The contamination rate of ^{127}I was calculated by the values of ^{127}I concentration in Table S1 (Supporting Information). Concentration of ^{127}I was in the range of 0.0159 – 4.15 μg L^{-1} (Table S1), consistent with earlier studies.[21] The level of ^{127}I contamination rate was dependent on each room and remained almost constant, regardless of sampling period. Averages of the ^{127}I contamination rates in units 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 type1, unrelated with chemistry, and can represent outdoor environmental air. As far the ^{127}I contamination rate, the air in room types2, 3, and 4 showed similar levels to outdoor environmental air. Earlier studies have suggested that iodine is present in the atmosphere at concentrations of 0.3 – 20 ng m^{-3}.[22,29] The amount of the ^{127}I collected in the trap solution was obviously smaller than the amount of ^{127}I in the whole room air as same as outdoor environmental air. The ^{127}I contamination rates in type5 rooms were significantly higher than the others. In these rooms, a larger amount of iodine reagents was used for chemical experiments than in the other rooms. The high ^{127}I contamination rates are attributed to the use of the reagents.

The contamination rate of ^{129}I

The measured ^{129}I contamination rates in the trap solutions in each room are plotted in Fig. 1. The $^{129}I/^{127}I$ ratios were $(2.0 \pm 0.6)\times 10^{-13}$ for the reagent blank, error means 1σ ($n = 6$), and $(2.0 - 5.6)\times 10^{-13}$ for procedure blanks. Our result of $^{129}I/^{127}I$ ratio $(2.0 \pm 0.6)\times 10^{-13}$ agrees well with the reference value[24,25] in the iodine reagent (Orion ionplus®) found by AMS at MALT, which has been reported at around 1.7×10^{-13}. The contamination rates of ^{129}I were calculated by using the values of ^{129}I concentration in Table S1. The associated uncertainty values of ^{129}I contamination rates accompanying the analytical procedures were within the range from 2 to 60%. The ^{129}I contamination rates in the trap solutions were quite different among the rooms. In 2014, the ^{129}I contamination rates in the type1 rooms were consistent with environmental levels, with the obtained $(0.67 - 1.2)\times 10^4$ atoms cm^{-2} day^{-1}, the ^{129}I level of atmospheric fallout samples was $(0.92 - 31.3)\times 10^4$ atoms cm^{-2} day^{-1}; calculated from $(0.39 - 13.3)\times 10^{-2}$ mBq m^{-2} $month^{-1}$ recorded from 1986 to 2005,[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,[21] in Tsukuba, Japan. Although the ^{129}I contamination rates in C209, $(0.41 - 3.2)\times 10^4$ atoms cm^{-2} day^{-1}, were comparable with those in type1 rooms (environmental level), the ^{129}I contamination rates in C210, $(1.0 - 26)\times 10^4$ atoms cm^{-2} day^{-1},
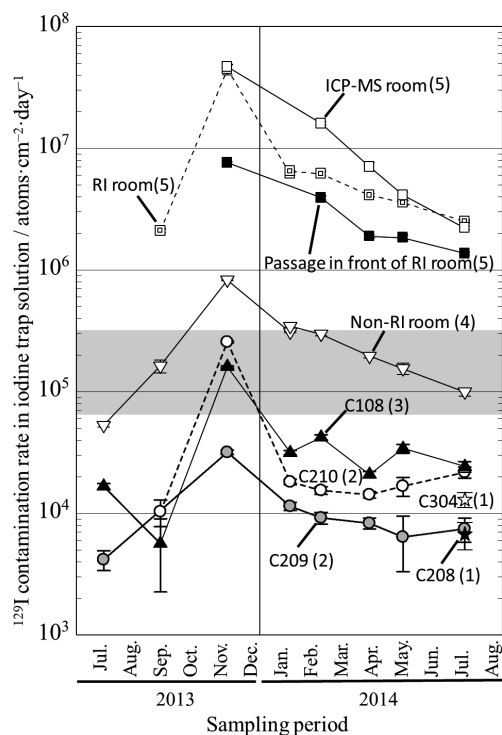
Fig. 1   Variation of the measured ^{129}I contamination rates in the trap solutions in each room. The symbol was plotted at a middle time of the sampling period on the vertical axis. Gray zones indicate background level from precipitation at Tsukuba.[21,30] The numbers in parentheses indicate room type.

Fig. 2   Variation of measured ^{129}I/^{127}I ratios in the trap solutions in 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.[21,30] The numbers in parentheses indicate room type.

and C108, (0.56 – 16)× 10^4 atoms cm^{-2} day^{-1}, were a little higher than those in type1 rooms. The ^{129}I contamination rates in the Non-RI room, (9.9 – 82)× 10^4 atoms cm^{-2} day^{-1}, was one order of magnitude higher than those in type1 rooms. Furthermore, the ^{129}I contamination rates in type5 rooms, (209 – 4431)× 10^4 atoms cm^{-2} day^{-1} for RI room, (221 – 4645)× 10^4 atoms cm^{-2} day^{-1} for ICP-MS room, and (136 – 756)× 10^4 atoms cm^{-2} day^{-1} for Passage in front of RI room, were two orders of magnitude higher than those for type1 rooms. The measured ^{129}I contamination rates pertain to the rooms' histories. As we expected, the rooms used for the treatment of samples with higher ^{129}I concentrations tended to have 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 ≈ 10^{16} atoms g^{-1}, ^{129}I/^{127}I ≥ 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 a part of them was diluted to certain concentrations in the RI room. Then, the diluted solutions were transferred to the ICP-MS room, where ICP-MS was performed for ^{127}I and ^{129}I, and stored there for approximately a month. Needless to say, we strictly followed the rules for the RI controlled area (*e.g.*, use of protective gloves and washing hands). However, the associated contamination was observed simultaneously in all the rooms, located outside of the RI controlled area, even in a different building. It was attributed to the active and volatile behavior of iodine being diffused by air and human movement. Fortunately, as shown in the variations of ^{129}I contamination rates in C209, C210, C108, Non-RI room, and RI room in
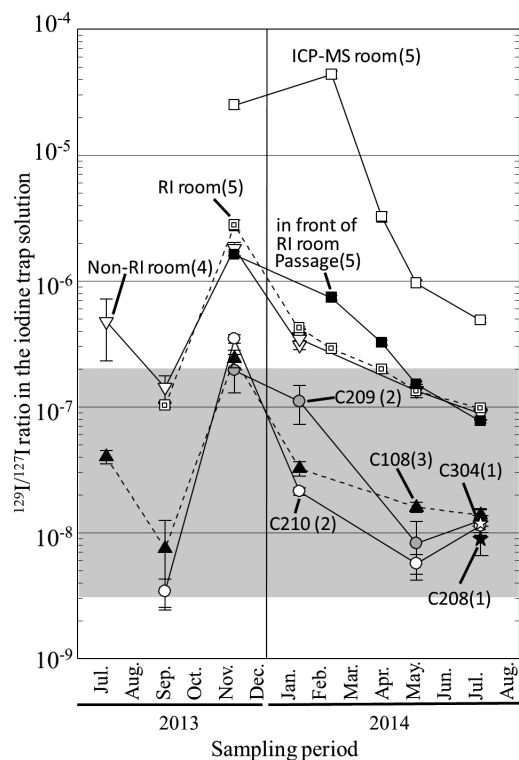
Fig. 1, the level of ^{129}I contamination decreased with time and returned to the level before the contamination occurred. It could have temporary effects across the laboratory.

The variations of ^{129}I/^{127}I ratios in the original trap solutions in each room are 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 also increased during that period and decreased with time. Then, the ^{129}I/^{127}I ratio returned to the level before the increase and became steady. The plot for the ICP-MS room appears different from those for the other two type5 rooms with similar histories. This room was extended recently (in 2013) inside the building. Therefore, ^{129}I contamination was strong, but ^{127}I contamination was not so advanced to the extent that the ^{129}I/^{127}I ratio in the ICP-MS room was higher than in other rooms in the same building. With the exception of the values obtained during 18 Nov. to 9 Dec. 2013, the ^{129}I/^{127}I ratio in the rooms with history types1 to 3 ranged around 2 × 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)× 10^{-8} recorded from 1986 to 2005[30] and (0.3 – 3.8)× 10^{-8} recorded from 2009 to 2010,[21] and after the FDNPP accident, (1.1 – 5.8)× 10^{-8} recorded in 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 the atmosphere was defined as the ^{129}I/^{127}I ratio obtained by AMS, according to the following equation;

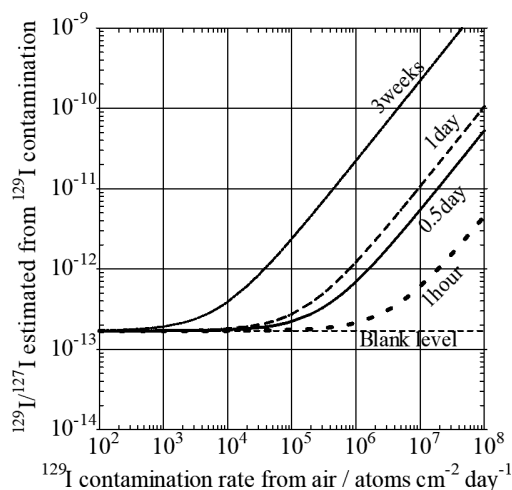Fig. 3   Variations of ^{129}I/^{127}I ratios in the procedure blank (Y-axis) adjusted for contributions of ^{129}I contamination (R) from room air estimated by Eq. (1), as a function of a variety of ^{129}I contamination rates (C, X-axis) at different working periods (T). The ratio of reagent blank is shown by a broken line, as "Blank level", 1.7×10^{-13}. Here, $W = 2$ mg and $A = 10$ cm^2.

$$R = {}^{129}\text{I}_{\text{precipitated from atmosphere}}(\text{atoms})/{}^{127}\text{I}_{\text{carrier}}(\text{atoms})$$
$$= (C \times A \times T)/(W \times \text{N}_\text{A}/127), \qquad (1)$$

where R is the contribution of the ^{129}I contamination from room air to the ^{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 period (day), W is the carrier weight (g) and N$_\text{A}$ is Avogadro constant. Here, within the results of the contamination rate of ^{127}I, the ^{127}I weight from the atmosphere (ng range) is much less than the weight of carrier iodine (mg range). Therefore, the amount of ^{127}I from the atmosphere is negligible for ^{129}I determination by AMS.

Figure 3 indicates variations of the ^{129}I/^{127}I ratio in the procedure blank estimated from ^{129}I contribution (R) of the various ^{129}I contamination rates by Eq. (1). Here, we plugged in 10 cm^2 ($e.g.$ beaker or centrifuge tube), 2 mg (general carrier weights for AMS), and 1 h, 0.5 day, 1 day and three weeks for parameters A, W, and T, respectively. The value from carrier reagent (reagent blank), 1.7×10^{-13} is shown by a broken line in Fig. 3. In the case of C108, for the indicated ^{129}I contamination rate among rooms that we usually used for AMS sample preparation, the contribution to ^{129}I/^{127}I ratio (R) was estimated to be 1.3×10^{-14} by Eq. (1) using ^{129}I contamination rate (C); 2.5×10^4 atoms cm^{-2} day^{-1} (average in the steady period from Fig. 1) and the working period (T); 0.5 day. Therefore, relative ^{129}I contribution to the ^{129}I/^{127}I atoms ratio of the procedure blank was 0.08, and estimated value of the ^{129}I/^{127}I ratio in the procedure blank is approximately 2×10^{-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 the 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 the RI room, R was estimated to be 2.2×10^{-12} by Eq. (1) using C; 4.1×10^6 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 the reagent blank. Therefore, it was found that a measured

value can be overestimated in the case of sample preparation in the RI room. During the high contamination period in November and December of 2013, by the same calculation, relative ^{129}I contribution to the ^{129}I/^{127}I atoms ratio of the 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. A measurement above the $(2.0 \pm 0.6) \times 10^{-13}$ background of the carrier reagent indicates atmospheric contamination of ^{129}I in the room or possibly some other reason, such as a long time taken for preparation or cross contamination or the other. However, we can estimate ^{129}I contamination risk roughly from the atmosphere using this method.

Conclusions

By the simple method using alkaline trap solutions, over one year, we monitored ^{127}I and ^{129}I contamination rates from the air in not only the three working rooms for AMS sample preparation, but also two rooms unrelated with chemistry and four rooms expected to show high ^{129}I contamination. It was found that the ^{127}I contamination with ng-order weight from the atmosphere was constant in 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 of the room. This fact implied that treated samples with higher ^{129}I concentrations tended to induce 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 the atmosphere for AMS was within the error range of AMS. Furthermore, a simultaneous high ^{129}I contamination event clearly showed variations of ^{129}I contamination rates in seven rooms. 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 discern the 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 in AMS sample preparation rooms by this method.

Supporting Information

Details of 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 are provided in Table S1. Chemical schemes are shown in Figs. S1 – S3. This material is available free of charge on the Web at http://www.jsac.or.jp/analsci/.

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