QA/QC Activities and Estimation of Uncertainty for Ultra-trace Analysis of Uranium and Plutonium in Safeguards Environmental Samples

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Japan Atomic Energy Agency (JAEA) has been conducting the environmental sample analysis, which is one of the IAEA's safeguards measures. In the analysis, isotope ratios of ultra-trace amounts of uranium and plutonium in samples taken from nuclear facilities are determined to detect undeclared nuclear materials and activities. In order to keep and enhance the reliability of the measurement results, validation of analytical methods and estimation of measurement uncertainty in such ultra-trace analysis should be properly done in accordance with worldwide standards. In this paper, the current activities on achievement of QA/QC (quality assurance and quality control) and estimation of measurement uncertainty in the ultra-trace analysis at a clean chemistry laboratory (CLEAR) of JAEA are reported.

1. Introduction

The International Atomic Energy Agency (IAEA) introduced the environmental sample analysis, which is to detect 'environmental signature' of undeclared nuclear materials (U and Pu) and activities, as one of safeguards measures based on 'Programme 93+2'.[1,2] In the environmental sample analysis, isotope ratios of U and Pu in samples taken from nuclear facilities and the related ones are analyzed at a clean chemistry laboratory. Main analytical methods are composed of 1) the bulk analysis providing information about the quantity and the average isotopic composition of each sample and 2) the particle analysis for U isotopic compositions of individual particles.

In Japan, the environmental sample analysis has been performed at Clean Laboratory for Environmental Analysis and Research (CLEAR) of Japan Atomic Energy Agency (JAEA).[3-7] Our aims of the analysis are 1) to maintain the independent verification scheme of the domestic safeguards system, 2) to contribute the IAEA as a member of NWAL (Network of Analytical Laboratories), and 3) to apply the developed techniques to other fields such as environmental science.

In order to find undeclared nuclear activities, ultra-trace analysis of U, Pu, minor actinides and long-lived fission products is necessary. Especially, the isotope ratios of U and Pu suggest their origin and related nuclear activities, *e.g.*, nuclear fuel, weapon, enrichment, reprocessing, natural grade or global fallout.

To keep and enhance the reliability of the measurement results, validation of analytical methods under cleanliness environment conditions at the level of worldwide standards and estimation of measurement uncertainty in such ultra-trace analysis should be reasonably done. This paper deals with the current activities and perspectives on achievement of QA/QC (quality assurance and quality control) and estimation of measurement uncertainty in the ultra-trace analysis of the safeguards environmental samples at CLEAR of JAEA.

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2. Achievement of QA/QC

The QA/QC system of the IAEA's NWAL should be based on the principles of worldwide standards. Main related international and national standards (or guidelines) in this field are as follows: 1) ISO/IEC 17025: 2005 [JIS Q 17025: 2005] (General requirements of the competence for testing and calibration laboratories), 2) ISO 14644 Series (Cleanrooms and associated controlled environments), and 3) ISO/IEC Guide 98 Part 3 (Guide to the expression of uncertainty in measurement [GUM: 1995]).

Based on the above standards, the clean chemistry laboratory, CLEAR, was designed, constructed and is being operated, and the environmental samples have been analyzed.[3-7] The samples are composed of non-radioactive ones and radioactive ones taken from a hot-cell facility and are, therefore, screened by radiometric techniques. On the other hand, the samples with less than 1 Bq order of U and Pu are acceptable for the ultra-trace analysis at CLEAR. The samples which contained much more radioactivity as hot-cell ones are analyzed at the radiochemistry facility, NUCEF (Nuclear Fuel Cycle Safety Engineering Research Facility) of JAEA.

The QA/QC and analytical procedures are systematized as 'Manuals on safeguards environmental sample analysis and quality assurance' as shown below:
1) Outline of analysis and quality control,
2) Sample receipt, storage and screening technique,
3) Bulk analysis for non-radioactive samples by ICP-MS (inductively coupled plasma mass spectrometry),
4) Hot-cell sample analysis for radioactive samples by TIMS (thermal ionization mass spectrometry),
5) Particle analysis by SIMS (secondary ion mass spectrometry) method,
6) Particle analysis by FT (fission track)-TIMS method, and
7) Cleanliness control.

In the QA/QC system of such a clean chemistry laboratory, in addition to the documentation and document control, a number of important requirements such as organization, management system, accommodation and environmental conditions, analytical methods and method validation, measurement traceability, *etc.* are contained as management and technical require-

ments. Here, activities on control of cleanliness environment and validation of methods are illustrated as examples of QA/QC achievement.

2.1. Control of cleanliness environment. The CLEAR is a key laboratory in Japan to accomplish the environmental sample analysis,[7] and consists of administration building and analytical building. Figure 1 shows the layout of analytical building and the classification of air cleanliness for each cleanroom area.

The cleanroom area is controlled as cleanliness environment at positive pressure mainly according to ISO 14644 Series and the manual of cleanliness control, while hot area except the cleanroom area is maintained at negative pressure according to the domestic regulation and JAEA's rules. Table 1 summarizes periodic cleanliness control activities in the analytical building prescribed in the manual of cleanliness control.

Usually no particle were detected in every hood and bench with ISO class 4, the upper limit of which is 1,020 particle/m^3 for the particles of more than 0.3 μm in diameter. As an example of cleanliness checks, Figure 2 shows monitoring results of airborne particle concentration in some cleanrooms. The cleanliness was unfailingly maintained at the respective classes.
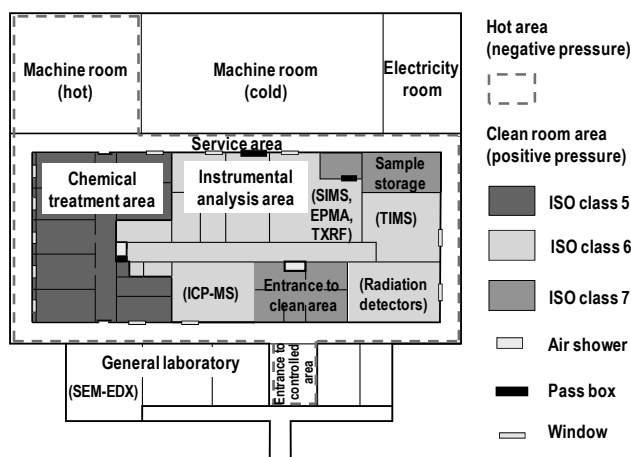

Figure 1. Layout of the analytical building of CLEAR.

TABLE 1: Cleanliness control activities in the analytical building of CLEAR

Daily inspection
- Cleanliness check
  1. Hoods (ISO Class 4) and benches (ISO Class 4–5): 1/month
  2. Cleanrooms (ISO Class 5–7): 1/month
  3. Air showers and pass boxes (ISO Class 5–6): if necessary
- Area blank measurement: 1/month
- Pressure difference: 1/day
- Temperature (23±1 °C [21±1 °C in winter]): 1/day
- Humidity (50± 5 % [45±5 % in winter]): 1/day

Annual inspection
- Air flow rate in hoods and benches: 1/year, FFU: 1/ 2 years
- Air flow direction: whenever necessary
- Air flow visualization: 1/year
- Filter leakage in hoods and benches: 1/year, FFU: 1/ 2 years
- Cleanliness recovery characteristics: 1/year

Cleaning
- Cleanrooms (ISO Class 5–7): 1/year
- Plenum chambers (ISO Class 5–7): 1/year

Area blank at several points in the cleanroom area is measured to check contamination of U, Th, Pb and other elements of interests. No significant contamination has ever been observed. Cleanliness recovery characteristics of the cleanrooms have been yearly examined. After restarting the fan-filter units (FFU), the cleanliness was immediately recovered within a few minutes.[4] Due to not only excellent designing of a clean chemistry laboratory but also the continuous activities of cleanliness control, the cleanliness environment at CLEAR has been successfully maintained.

2.2. Validation of methods. The analytical methods and procedures suitable for various types of the environmental samples have been developed.[8-27] The methods and procedures were validated, for example, by the use of reference materials certified by NBL (New Brunswick Laboratory) such as CRM111A (^{233}U), CRM130 (^{242}Pu), CRM U015 (U) and SRM947 (Pu) as spikes and isotopic standards.

JAEA has been participating in inter-laboratory comparisons operated by IRMM (Institute for Reference Materials and Measurements),[28] such as NUSIMEP (Nuclear Signatures Interlaboratory Measurement Evaluation Programme) and REIMEP (Regular European Interlaboratory Measurement Evaluation Programme),[29] for validation of the developed bulk and particle analytical methods. The JAEA's measurement results and capabilities were proved to be excellent in the comparisons.

The IAEA has crosschecked the analytical results and done blind tests using QC samples. The methods and results have been discussed and evaluated every year at the IAEA technical meetings, which are composed of the IAEA and NWAL members. The JAEA's methods and results have been evaluated to be reliable and accurate until now.

In addition, process blank of U and Pu must be exactly analyzed to correct the measured values. The blank amounts in the bulk analysis at CLEAR were approximately 10 pg of natural U and 0 fg of Pu. It was found to be maintained at the extremely low level even now. Consequently, the effect of blank on isotope ratio measurement at CLEAR was negligible.

3. Estimation of uncertainty

The IAEA recommends the typical accuracies in the analysis for the environmental samples with expected characteristics. The accuracies for each instrument of mass spectrometry for the bulk analysis and the particle analysis at JAEA were summarized in Table 2, together with the IAEA-expected values in the table foodnote.

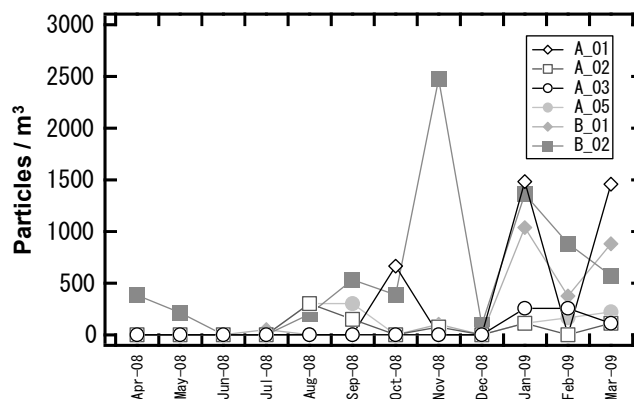The accuracies obtained for the respective instruments were satisfactory, compared with those recommended by IAEA

Figure 2. Example of cleanliness check results of airborne particle concentration in some cleanrooms of the chemistry area (A 01-05: ISO class 5 [upper limit of 0.3 μm sized particles; 10,200/m^3] and instrument area (B 01, 02: ISO class 6 [upper limit; 102,000/m^3]).

TABLE 2: Accuracies obtained for each instrument of mass spectrometry at JAEA

Instruments (Techniques)	Measured values	Range	Relative accuracy (2σ) [a]
ICP-MS [ELEMENT]	U amount	10^{-11}–10^{-6} g	0.5 % (at 5 ng natural U) [b]
(CLEAR/bulk by IDM)	Pu amount	10^{-14}–10^{-10} g	5 % (at 1 pg) [c]
TIMS [MAT262]	U amount	10^{-9}–10^{-4} g	1 % (at 4 ng natural U) [b]
(NUCEF/ hot-cell sample by IDM)	Pu amount	10^{-13}–10^{-7} g	1 % (at 1 ng) [c]
ICP-MS [ELEMENT]	$^{235}U/^{238}U$ ratio	2×10^{-6}–10^{2}	0.6 % (at 500 pg/mL natural U) [d]
(CLEAR/bulk)	$^{240}Pu/^{239}Pu$ ratio	10^{-3}–10^{2}	1 % (at 5 pg/ml SRM947) [e]
TIMS [MAT262]	$^{235}U/^{238}U$ ratio	10^{-6}–10^{2}	≤1 % (at 10 ng natural U) [d]
(NUCEF/hot-cell sample)	$^{240}Pu/^{239}Pu$ ratio	10^{-4}–10^{2}	≤1 % (at 100 pg Pu) [e]
SIMS [Cameca 6f]	$^{235}U/^{238}U$ ratio	10^{-5}–10^{2}	≤10 % (at 10 pg natural U) [f]
(CLEAR/particle by SIMS)	$^{240}Pu/^{239}Pu$ ratio	–	–
TIMS [TRITON]	$^{235}U/^{238}U$ ratio	10^{-6}–10^{2}	≤5 % (at 10 pg natural U) [g]
(CLEAR/particle by FT-TIMS)	$^{240}Pu/^{239}Pu$ ratio	10^{-4}–10^{2}	≤5 % (at 1 pg SRM947)

a: Corresponding to confidence level ≈ 95%, b: typical accuracy expected by IAEA; ≤20 % for 10 ng of U, c: ≤10 % for 5 pg of Pu, d: ≤1 % for 10 ng of U, e: ≤10 % for 5 pg of Pu, f: ≤10 % for 1 μm particle of UO_2 (~5 pg of U), g: ≤1 % for 1 μm particle of UO_2.

TABLE 3: Accuracies in radioactivity measurements

Methods	Facility	Range	Relative accuracy
γ-ray spectrometry	CLEAR	10^{-3}–10^{5} Bq	5–10 % (at 1 kBq ^{137}Cs)
	NUCEF	10^{1}–10^{9} Bq	5–10 % (at 10 kBq ^{137}Cs)
α-ray spectrometry	CLEAR	10^{-4}–10^{4} Bq	2–5 % (at 3 kBq ^{244}Cm)
	NUCEF	10^{-3}–10^{6} Bq	2–5 % (at 10 kBq ^{241}Am)

Expected γ-activity range by IAEA: 0.01 to 10^{8} Bq/swipe, expected relative accuracy: ≤ 10 %.

TABLE 4: Components of standard uncertainty in bulk analysis at CLEAR

Type A (The statistical analysis)

Standard deviation of the mean of each isotope ratio measured value
Correction of mass bias ($^{235}U/^{238}U$ ≈ 0.5 %)
Correction of blank (quantity: ≤1%, ratio: negligible)
Correction of tailing ($^{236}U/^{238}U$ ≈ 2×10^{-6})
^{236}U correction by UH evaluation (≈ 4×10^{-5})

Type B (Means other than the statistical analysis)

Data sheet of reference materials (spike and mass bias) [<1%]
Weighting (negligible)
Evaporating (spike solution ≈ -0.03% per month)
Adsorption and leaching out (negligible)

except for the particle analysis by TIMS, however, it is difficult to simply compare between them because of different analytical conditions.

The accuracies in radioactivity measurements for screening and/or analyzing minor actinides, fission and activation products by α- and γ-ray spectrometry were also roughly obtained (see Table 3). They mainly depended on accuracy in the data sheet of radioisotope reference sources such as ^{137}Cs, ^{241}Am and ^{244}Cm.

3.1. Uncertainty in bulk analysis. Concentrations of U and Pu and their isotope ratios are determined with IDM (isotope dilution method) by mass spectrometry. The traceability of the results to the SI (Systeme Internationale) units of measurement is established mainly by utilizing the certified reference materials and the measured values are properly corrected. Each standard uncertainty is evaluated by separating Type A and B in accordance with the GUM. Table 4 summarizes components of the standard uncertainty in bulk analysis at CLEAR.

Usually, the standard deviation of the mean of each isotope ratio measured value in Type A is main component and the others are minor. Uncertainty in weighing was negligible, even if it was overevalued. Change in the concentration of the spike solution due to evaporation was corrected by weighting it before use. Adsorption of Pu into various vessels and leaching of U from the vassels were also examined to be negligible because the vessels of high quality were selected and used.

Table 5 shows a representative example of the uncertainty evaluation in U isotope ratio measurement of a blank swipe sample (swipe cotton cloth: TexWipe-304), which currently used in the environmental analysis of the IAEA and contains a few ng of natural U.[30] In every case, the standard deviation of the mean of each isotope ratio measured value was main component. In $^{236}U/^{238}U$ ratio analysis, correction of tailing from ^{238}U peak to ^{236}U peak was serious component. The relative expanded uncertainty of the corrected $^{236}U/^{238}U$ ratio was over 100 %.

3.2. Uncertainty in particle analysis. Isotope ratios of U in a single particle in each sample are determined by SIMS and FT-TIMS methods. The standard uncertainty in the SIMS particle analysis has been reported according to the IAEA procedures. In this paper, components of the uncertainty is estimated in the same way as the bulk analysis (see Table 6).

The standard deviation of the mean of each isotope ratio measured value in Type A is also main component and the others are minor. The blank amounts in the particle analysis were negligible at CLEAR. In the SIMS method, uncertainty in ^{236}U correction by UH evaluation is nonnegligible component of the uncertainty, and determination of $^{240}Pu/^{239}Pu$ ratio was exceedingly difficult due to the ^{238}UH formation.

At present, investigation is focused on minor isotope ratios of U and/or Pu in sub-micrometer sized particles by the FT-TIMS method. In bulk analysis, the interferance of alkali metals as well as polyatomic ions, such as IrAr, PtAr, AuAr, PbO_2, ReO_3, WO_3, *etc.*, on the isotope ratio measurements of U and Pu have been well examined, and chemical separation was

TABLE 5: A representative example of uncertainty evaluation in U isotope ratio measurement of a blank swipe sample (Instrument: ICP-MS)

Measured values	Type of uncertainties	Components or sources	Values	Uncertainties (Relative values)	Relative Contribution
$^{235}U/^{238}U$ ratio	A	$^{235}U/^{238}U$ ratio of sample	7.233×10^{-3}	2.3×10^{-5} (0.32 %) [c]	95 %
		$^{235}U/^{238}U$ ratio of reference [a]	1.54327×10^{-2}	8.6×10^{-6} (0.056 %) [c]	2.8 %
	B	Number of ^{235}U atoms of reference [b]	1.53230×10^{-2}	7.5×10^{-6} (0.049 %) [c]	2.2 %
		Number of ^{238}U atoms of reference [b]	9.84430×10^{-1}	1.0×10^{-5} (0.00001%) [c]	0.0 %
		Weighting, evaporating, *etc.*	—	—	(Negligible)
		Corrected $^{235}U/^{238}U$ ratio	7.295×10^{-3}	2.4×10^{-5} (0.33 %) [d]	—
$^{234}U/^{238}U$ ratio	A	$^{234}U/^{238}U$ ratio of sample	7.76×10^{-5}	5.3×10^{-6} (6.8 %) [c]	100 %
		$^{235}U/^{238}U$ ratio of reference [a]	1.54327×10^{-2}	8.6×10^{-6} (0.056 %) [c]	0.0 %
	B	Number of U atoms of reference [b]	—	—	0.0 %
		Weighting, evaporating, *etc.*	—	—	(Negligible)
		Corrected $^{234}U/^{238}U$ ratio	7.85×10^{-5}	5.4×10^{-6} (6.9 %) [d]	—
$^{236}U/^{238}U$ ratio	A	$^{236}U/^{238}U$ ratio of sample	2.80×10^{-6}	5.0×10^{-7} (18 %) [c]	34 %
		$^{235}U/^{238}U$ ratio of reference [a]	1.54327×10^{-2}	8.6×10^{-6} (0.056 %) [c]	0.0 %
		Correction of UH evaluation	3.33×10^{-5}	2.5×10^{-6} (7.5 %) [c]	0.0 %
		Tailing from ^{238}U to ^{236}U peak	1.80×10^{-6}	7.0×10^{-7} (39 %) [c]	66 %
	B	Number of U atoms of reference [b]	—	—	0.0 %
		Weighting, evaporating, *etc.*	—	—	(Negligible)
		Corrected $^{236}U/^{238}U$ ratio	7.6×10^{-7}	8.7×10^{-7} (114 %) [d]	—

a: Mass bias correction, b: data sheet of reference, c: standard uncertainty, d: expanded uncertainty (coverage factor: 2, confidence level: 95%).

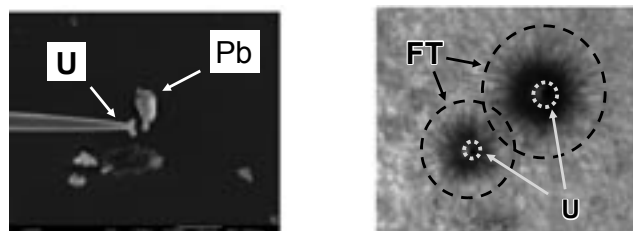**TABLE 6: Components of standard uncertainty in particle analysis**

Type A (The statistical analysis)
Standard deviation of the mean of each isotope ratio measured value
Mass bias correction ($^{235}U/^{238}U \approx 1$ %)
Tailing correction (negligible)
[TIMS: $^{237}U/^{238}U \approx 2\times10^{-6}$]
^{236}U correction by ^{235}UH
($\approx 1\times10^{-4}$ by SIMS, negligible by TIMS)
Type B (Means other than the statistical analysis)
Data sheet of reference materials [< 1%]

Figure 3. Picking up a particle containing U by observing SEM-EDX image (left) and identification of particles containing fissile materials by observing FT (right).

applied to reduce the interference.[9,10] On the other hand, it is difficult to complement the isotope ratio measurements in the particle analysis, because it is performed without chemical separation in principle. In future, besides the UH evaluation, interference of polyatomic ions with the minor isotope ratios should be investigated.

In the particle analysis, in addition, there is another serious problem of "Mixing", which means simultaneous measurement of plural particles of different isotope ratios, resulting in an intermediate value of the measured ratio. In order to overcome the problem in SIMS method, we developed a method to pick up individual particle containing U or fissile materials by micro-manipulation in SEM-EDX (scanning electron microscope-energy dispersive X-ray analysis) (see Figure 3 left).[14-16] For the FT-TIMS method, it is easy to observe FT by an optical microscope and to collect the particles separately (see Figure 3 right).[21-23] This method enables us to selectively detect highly enriched U particles, which is critical in the nuclear safeguards.[25]

4. Conclusions

Cleanliness control of the CLEAR and validation of analytical methods were successfully achieved. The uncertainty in the environmental bulk and particle analyses has been routinely evaluated. On the other hand, the difficulties in the particle analysis should be solved in near future. As a result, achievement of QA/QC and estimation of measurement uncertainty in ultra-trace analysis of U and Pu are at the level of worldwide standards, which properly keeps and enhances the reliability of the measurement results at CLEAR.

Further pursuit of sensitivity and reliability will be necessary for development of advanced techniques of determining the isotope ratios of U and Pu. Anytime, efforts are required to estimate potential sources of the uncertainty and to evaluate its amount.

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