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GHSI Emergency Radionuclide Bioassay Laboratory Network - Summary of the Second Exercise

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Abstract

The Global Health Security Initiative (GHSI) established a laboratory network within the GHSI community to develop collective surge capacity for radionuclide bioassay in response to a radiological or nuclear emergency as a means of enhancing response capability, health outcomes and community resilience. GHSI partners conducted an exercise in collaboration with the WHO REMPAN (Radiation Emergency Medical Preparedness and Assistance Network) and the IAEA RANET (Response and Assistance Network), to test the participating laboratories (18) for their capabilities in *in vitro* assay of biological samples, using a urine sample spiked with multiple high-risk radionuclides (^{90}Sr, ^{106}Ru, ^{137}Cs, and ^{239}Pu). Laboratories were required to submit their reports within 72 hours following receipt of the sample, using a pre-formatted template, on the procedures, methods and techniques used to identify and quantify the radionuclides in the sample, as well as the bioassay results with a 95% confidence interval. All of the participating laboratories

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identified and measured all or some of the radionuclides in the sample. However, gaps were identified in both the procedures used to assay multiple radionuclides in one sample, as well as in the methods or techniques used to assay specific radionuclides in urine. Two third of the participating laboratories had difficulties in determining all the radionuclides in the sample. Results from this exercise indicate that challenges remain with respect to ensuring that results are delivered in a timely, consistent and reliable manner to support medical interventions. Laboratories within the networks are encouraged to work together to develop and maintain collective capabilities and capacity for emergency bioassay, which is an important component of radiation emergency response.

Keywords

nuclear emergency; radiological emergency; radioactive contamination; radionuclide bioassay; internal dosimetry

Introduction

Following a radiological or nuclear emergency, workers, first responders and the public may be internally contaminated with the radionuclide(s) involved. Rapid assessment of internal contamination, through *in vitro* or *in vivo* bioassay, provides timely information for medical intervention, if necessary, and provides assurance to those who do not require further examination. Accordingly, emergency radionuclide bioassay capacity is essential to support emergency response. In a large-scale emergency, it is possible that laboratories from other regions or other countries may be called upon for assistance.

The Global Health Security Initiative (GHSI) is an informal network of countries formed in 2001 to ensure health-sector exchange and coordination of practices in confronting risks to global health posed by chemical, biological and radio-nuclear threats, as well as by pandemic influenza[(1)]. The member countries/organizations of the GHSI are Canada, France, Germany, Italy, Japan, Mexico, the United Kingdom, the United States and the European Commission. The World Health Organization (WHO) is a technical advisor to the GHSI. As part of the GHSI partnership, an annual meeting of Health Ministers is held to foster dialogue on topical policy issues and promote collaboration. Other initiatives involving senior health officials as well as policy, technical and scientific personnel take place on a regular basis, focused on risk management; communications; chemical events; radio-nuclear threats; pandemic influenza; and global laboratory cooperation.

The GHSI Radio-Nuclear Threats Working Group (RNWG) was created to facilitate sharing and collaboration on policies and capability development to enhance public health preparedness and response to radiological and nuclear threats. As a result of discussions and consultations, the RNWG established an informal laboratory network to improve collective capabilities and capacity for radionuclide bioassay within the GHSI community. Within this network, laboratories can share their expertise through training activities, exercise their preparedness through intercomparisons, develop new capabilities through collaborative R&D, and assist in bioassay analysis when additional laboratories are required following an emergency.

In 2013, the network laboratories were surveyed on their current capabilities in emergency radionuclide bioassay and the technological and operational gaps they had identified in this area. Based on the survey results, the RNWG decided to conduct two exercises. The first exercise was organized in 2014 to test the participating laboratories for their response capabilities in assaying a single radionuclide (^{241}Am) in a urine sample, performing internal dose assessment, and providing advice on medical intervention when necessary. Results for the first exercise have been published [(2)].

The second exercise, organized in early 2016 and reported in this paper, is to test the participating laboratories (18) for their response capabilities in assaying multiple radionuclides in a single urine sample, focusing on the procedures and methods/techniques used and the results obtained by the participating laboratories. This exercise was in collaboration with the WHO REMPAN (Radiation Emergency Medical Preparedness and Assistance Network, World Health Organization) [(3)] and the IAEA RANET (Response and Assistance Network, International Atomic Energy Agency) [(4)]. Some laboratories from the REMPAN collaborating centers and liaison institutions and the ones that had registered their bioassay capabilities in the RANET database participated in this exercise, together with laboratories in the GHSI network.

Methods and Materials

Exercise Design

A scenario (unspecified) based on a severe nuclear power plant accident was adopted, with a simplified selection of radionuclides for the purpose of this exercise. The four selected radionuclides (^{90}Sr, ^{106}Ru, ^{137}Cs and ^{239}Pu) represent significant dose contributors potentially released from a nuclear power plant accident; they also serve as good candidates to test the participating laboratories on their bioassay capabilities for mixed α, β, and γ emitters. The activity ratios of the four radionuclides (^{106}Ru: ^{137}Cs: ^{90}Sr: ^{239}Pu = 1: 1.16: 0.137: 1.78×10^{-4}) are referenced to the source term of the Chernobyl accident [(5)] although in another accident these may be very different as a result of many factors, such as the design of the reactor and the technologies used to reduce the source term. The ratios of these radionuclides in the intake by an individual would also be very different as the dispersion and deposition of these radionuclides may be controlled by different parameters. Nevertheless, these ratios were used as a reference when selecting the intake activities of the radionuclides in this project.

NCRP 161 [(6)] introduces a new operational quantity, the Clinical Decision Guide (CDG), for physicians to consider the need for medical treatment of internal contamination. The CDG value is the intake of a radionuclide that satisfies both the stochastic health effect criterion (a 50 y committed effective dose (CED) of 0.25 Sv for adults) and the deterministic health effect criterion (the 30 d RBE-weighted absorbed dose values of 0.25 Gy-Eq to bone marrow or 1 Gy-Eq to the lungs, for adults). For the four radionuclides considered in this project, the stochastic health effect criterion applies. NCRP 161 further recommends that for inhalation exposure, a particle size of 5 μm AMAD (activity median aerodynamic diameter) may be used in the derivation of the CDGs as it is judged to be a reasonable default particle size because intakes of clinical significance are more likely to occur near the point of release

where the sizes of airborne particles are usually the largest and the concentrations of airborne radionuclides are usually highest [6].

Among the four radionuclides, ^{106}Ru would give the highest CED if the release scenario was the same as the Chernobyl accident. For a CED of 0.25 Sv, using the default inhalation solubility (type M) for ^{106}Ru according to ICRP 71[7] and calculating the intake and bioassay quantities using IMBA Plus® (version 4.0.36, provided by ACJ & Associates, Inc., 129 Patton Street, Richland, WA, USA), the calculated intake is 1.50×10^7 Bq and the calculated urine excretion on Day 7 is 1.46×10^4 Bq (or 2.28×10^3 Bq in 250 mL, using default daily urine excretion of 1.6 L recommended by ICRP). Based on the intake activity of ^{106}Ru and the source term ratios discussed above, the calculated intakes for ^{137}Cs (type F), ^{90}Sr (Type M) and ^{239}Pu (Type M) are 1.74×10^7 Bq, 2.06×10^6 Bq and 2.67×10^3 Bq, respectively. Urine excretions on Day 7 for these three radionuclides would be 6.64×10^4 Bq (or 1.04×10^4 Bq in 250 mL), 2.89×10^3 Bq (or 4.52×10^2 Bq in 250 mL), and 65.1 mBq (or 10.2 mBq in 250 mL), respectively. Table 1 lists the activities (Bq) spiked in a 250 mL urine sample provided to each participating laboratory and their concentrations (Bq/L). The spiked activities are corresponding to about 1% of the above calculated activities for ^{137}Cs, ^{90}Sr and ^{106}Ru, and about 100% of that for ^{239}Pu in a 250 mL urine sample collected on Day 7 following exposure, taking into consideration both the objectives of this exercise and the technical challenges in assaying each radionuclide.

Sample Preparation and Distribution

Blank urine was collected from healthy unexposed individuals, pooled, preserved with 1% HCl, and spiked with the four radionuclides: ^{90}Sr (SRM 4919I, NIST, Gaithersburg, MD, USA), ^{106}Ru and ^{239}Pu (Lot 1847-55-1 and Lot 1443-58-2, respectively, Eckert Ziegler Isotope Products, Valencia, CA, USA), and ^{137}Cs (Lot S0/35/57, Amersham Laboratories, Buckinghamshire, UK), following the standard procedure of the National Calibration Reference Center for Bioassay and *In Vivo* Monitoring, Health Canada, which is certified to the International Organization for Standardization (ISO) 9001:2008 standard [8]. The spiked urine sample was then divided into 250 mL aliquots; one was sent to each participating laboratory by a commercial courier without freezing. Laboratories were required to report results within 72 hours following receipt of the sample. The 250 mL sample size was chosen to enable all of the laboratories to use their existing analytical methods, however, in an emergency situation, a laboratory would only expect to receive about 30 to 100 mL of urine on average based on past public health emergency responses (CDC personal communication).

Results and Discussions

Response to Reporting Schedule

A message was sent to the participating laboratories immediately after the samples were picked up by the courier. The majority of the laboratories received the samples within three days. A small number of them experienced a short delay mainly due to customs clearance. One laboratory ultimately withdrew from this exercise after discovering that customs clearance would take up to a month.

Overall, 15 of the participating laboratories submitted their reports within 72 hours of receiving the sample (the required reporting schedule). Three laboratories started late as a result of a scheduling conflict with other work commitments or instrument breakdown, but submitted their reports within 72 hours. Considering the fact that the radionuclides in the sample are unknown to the participating laboratories, identification and quantification of different radionuclides require different methods and techniques, and the measurement of radionuclides at low levels takes time, reporting the results within the required reporting schedule of 72 hours is acceptable. However, it is worthwhile to note that during a real emergency, when faced with hundreds to thousands of samples, laboratories are expected to have very short turnaround time for sample analysis and reporting so to support decisions on medical intervention.

Reported Procedures for Screening the Sample

The laboratories did not know what radionuclides were spiked in the sample, or the activity levels. The participating laboratories were only notified that the urine sample simulates a collection from an individual who was contaminated during a severe nuclear power plant accident and that the selection of radionuclides was simplified for the purpose of this exercise.

Screening the sample using available instrumentation and procedures before assaying specific radionuclides can help identify certain radionuclides in the sample, for example, if beta/alpha emitters are present, or even provide qualitative evaluation on the activity levels of certain radionuclides, depending on the instrumentation and procedures used. Eleven out of the 18 participating laboratories screened the samples before performing assays for specific radionuclides. All of them screened using gamma spectrometry using the whole or a fraction of the sample received (L02, L03, L05, L06, L08, L09, L10, L13, L17, L18, and L19), while four of them (L02, L03, L09, and L10) also screened for gross beta/alpha using liquid scintillation counting. Results from such screening can help in selecting the methods/techniques for assaying specific radionuclides, determining the size of sub-samples for different analyses, and allocating the time for sample preparation and measurements.

Reported Methods and Techniques for Assaying Individual Radionuclides

Splitting the sample received (250 mL) into sub-samples for the assays of different radionuclides using different methods/techniques is the common approach of many participating laboratories. This allows the assays for different radionuclides to be carried out at the same time and ultimately helps reduce the sample turnaround time.

High resolution gamma spectrometry was used for assaying both ^{137}Cs (photon peak of 662 keV for ^{137m}Ba) and ^{106}Ru (photon peak of 622 keV for ^{106}Rh) in the sample by 15 out of the 18 participating laboratories, with the volume of sub-sample varying from 10 mL to 250 mL and the counting time varying from 15 min to 64 hours. All 15 laboratories identified and quantified ^{137}Cs in the sample, but three of them missed ^{106}Ru. This was mainly caused by the small volume of sub-sample and/or the short counting time used, as the photon peak area of ^{106}Rh (representing ^{106}Ru) is much smaller than that of ^{137}Cs. A larger volume and/or longer counting time would improve the identification and quantification of ^{106}Ru in

the sample. However, during a real emergency, the available volume of a sample may be small and the counting time may have to be short.

Assaying ^{90}Sr in the sample might be the most challenging task of this exercise. Only half of the 18 participating laboratories reported results for ^{90}Sr. Unless chemical separation is applied, direct assay using typical liquid scintillation counting would not tell the presence of ^{90}Sr (or its daughter ^{90}Y) in the sample as there are two other beta emitting radionuclides in the sample, ^{106}Ru and ^{137}Cs, whose activities are higher by a factor 4 and 20, respectively. Comparing the measurements for gamma activities and gross beta activities in the sample, especially when the counting time is short, would not help in identifying a pure beta emitter, in this case ^{90}Sr, as its activity is very low compared to that of other radionuclides. This was demonstrated by one laboratory. In this exercise, the responding laboratories separated ^{90}Sr or its daughter ^{90}Y from an aliquot of the urine sample following chemical treatments involving precipitation, digestion and separation using ion exchange, ion chromatography or extraction chromatography. ^{90}Sr was quantified either by measuring itself or the ingrowth of its daughter, ^{90}Y, using liquid scintillation counting, gas flow proportional counting, or Čherenkov counting.

Thirteen participating laboratories reported results for ^{239}Pu with one of them reporting a level of less than the detection limit of the method used (70 mBq/L). The measurement of ^{239}Pu in the sample was made either using inductively coupled plasma mass spectrometry (ICP-MS, L02) or using alpha spectrometry (the other 12 laboratories) following extensive sample preparation, which involves adding tracers to the sub-sample, precipitating the analyte using calcium (and magnesium for one laboratory) phosphate, decomposing the precipitate using HNO_3 (and H_2O_2 in some laboratories) with or without a microwave system, separating the analyte using anion exchange chromatography and/or solid phase extraction chromatography, preparing the source using lanthanide fluoride micro-precipitation or electrodeposition on stainless steel discs, and counting the source using alpha spectrometry. The volume of the sub-sample and the counting time used for the measurement of ^{239}Pu in the sample varied significantly among the laboratories, which resulted in a wide range of detection limits and measurement accuracy.

As described above, the measurements of ^{239}Pu and ^{90}Sr in the sample involved chemical separation steps. Tracers, such as ^{242}Pu for ^{239}Pu, and carrier, such as stable strontium for ^{90}Sr, were used by many of the participating laboratories. The use of tracer or carrier allows the monitoring of chemical recoveries and correction of the final assay results.

Five laboratories applied procedures for sequential separation of the radionuclides in the sample, using either an ion exchange column (L05, L08, L09) or a stack of solid phase extraction columns (L06, L12). A procedure with sequential separation capability allows the separation of multiple radionuclides in one sample, avoids splitting the sample for the assays of different radionuclides, saves time in sample preparation, and improves the quality of assay results.

ICP-MS is a rapidly growing technique for the measurement of trace elements. It has been applied to the measurements of radionuclides in human and environmental samples thanks to

its super sensitivity. As demonstrated by one laboratory (L02), ^{239}Pu in the urine sample can be quantified using only 1 mL of the sample, by ICP-MS in conjunction with solid phase extraction and an efficient sample introduction system. However, as the concentrations of the radionuclides involved in this exercise are very low, without appropriate sample preparation (for chemical purification) and specialized sample introduction to ICP-MS for enhanced ion transport efficiency, it would not be possible to measure them using ICP-MS due to insufficient limits of detection (L01, L08, L16).

It is worthwhile to note that many laboratories reported their efforts in identifying and quantifying many other radionuclides, namely ^{3}H, ^{14}C, ^{89}Sr, ^{238}U, ^{238}Pu, ^{241}Am, ^{242}Cm, and ^{244}Cm, which are not involved in this exercise but are very relevant to the scenario communicated to the laboratories. In a realistic severe nuclear power plant accident, some of them might be present in the bioassay samples collected from the affected individuals.

Reported Bioassay Results

Table 2 and Figures 1–4 present the bioassay results reported by the participating laboratories with uncertainties at 95% confidence interval (CI). The reported concentrations of the radionuclides in the sample are quite close to the spiked levels (Table 1) in this exercise, with a small number of exceptions (e.g. ^{137}Cs reported by L16 and ^{90}Sr reported by L09). The reported uncertainties for all of the four radionuclides vary significantly. This is caused mainly by the differences in the volume of the sub-sample, assay method or technique, and the counting time used by the participating laboratories. The methods used to estimate assay uncertainties by the laboratories vary, but most of them follow the established methods for uncertainty estimation and propagation, published as national or international standards. It is worthwhile to note that there are no published acceptance criteria for emergency bioassay. International organizations, such as the ISO (International Organization for Standardization), may consider the feasibility of developing such criteria.

As discussed above, ^{90}Sr, ^{106}Ru, ^{137}Cs and ^{239}Pu were selected for this exercise as they would be significant dose contributors potentially released from a severe nuclear power plant accident and are good candidates to test the participating laboratories on their bioassay capabilities for mixed α, β, and γ emitters in one sample. In the stock solutions used to spike the urine sample, ^{90}Sr and ^{90}Y, and ^{106}Ru and ^{106}Rh, have reached equilibrium, respectively. Three laboratories (L03, L08, and L13) reported results for ^{106}Ru as ^{106}Ru/^{106}Rh. Four laboratories (L06, L08, L12, and L13) reported ^{239}Pu as ^{239,240}Pu because the energy resolution for alpha spectrometry cannot differentiate the signal from ^{239}Pu and that from ^{240}Pu. One laboratory (L13) reported ^{90}Sr as ^{89,90}Sr as the chemistry used to extract ^{90}Sr from the urine sample and the measurement using gas flow proportional counting could not differentiate the signal of ^{90}Sr from that of ^{89}Sr, another important radionuclide from the nuclear fission process.

Gaps Identified for Assaying Multiple Radionuclides in One Sample

Following an RN emergency, source term assessment, environmental monitoring or other investigations would inform the key radionuclides released from the incident. However, sometimes the radionuclides information may not be immediately available. Laboratories

performing bioassay may need to screen the samples to identify the radionuclides involved, select the methods/techniques for assaying each of them, determine the sizes of sub-samples for different analyses, and allocate time for sample preparation and measurements. In this exercise, eight of the participating laboratories did not report their efforts in sample screening; they only reported the assays of specific radionuclides in the sample.

Only six of the 19 participating laboratories reported results for all of the four radionuclides. The other laboratories reported results for some or none of them. These laboratories may not have the methods and techniques to identify and/or quantify all of the radionuclides (α, β, and γ emitters); some laboratories may only be equipped with gamma spectrometers while some others may only be equipped with ICP-MS.

This exercise reveals that, generally, participating laboratories have the technical capacity to assess internal contamination to support effective public health and medical response to a nuclear emergency, although for some of them, their capacity is quite limited. To be useful in a response, in addition to being accurate, assessments must also be timely and sometimes need to be done with a smaller volume of sample (say, 30 to 100 mL). In the event of a large-scale emergency that overwhelms national bioassay capacity, delivering a timely result will require efficient international coordination among laboratories, including sharing samples and delivering results. Although efforts within GHSI, WHO, and IAEA have made notable progress towards establishing international laboratory networks to provide surge capacity and related assistance to support an emergency response, results from this exercise indicate that challenges remain to ensuring that results are delivered in a timely, consistent and reliable manner to support medical interventions.

Recommendations

The GHSI Laboratory Network for Emergency Radionuclide Bioassay has been working on identifying the gaps and developing capabilities and surge capacity for responding to a radiological or nuclear (RN) emergency. This informal laboratory network is complementary to the assistance capabilities established through RANET (IAEA) and REMPAN (WHO). It is recommended that the three networks further strengthen their collaborations and leverage their efforts in developing the global capabilities and capacity for emergency radionuclide bioassay, including: (1). Organizing a technical workshop to facilitate exchanges and learning among the laboratories; (2). Initiating a laboratory hands-on training program among laboratories; (3). Developing a practical plan for coordinating sample sharing and analysis, and integrating this plan into national emergency preparedness and response. These recommendations are those of the GHSI laboratory network and based on information gathered from the two exercises. These recommendations do not represent one particular agency.

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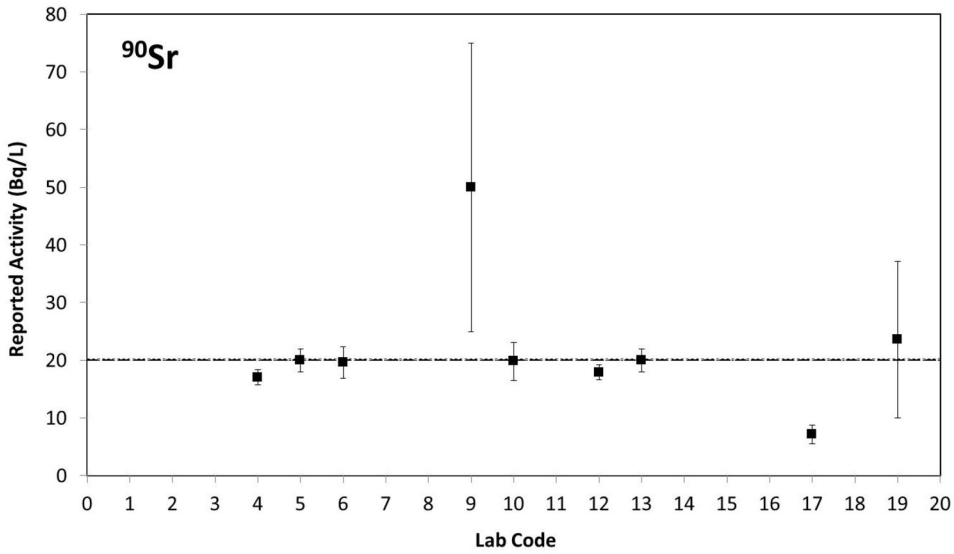

Figure 1.

Distribution of the reported results for ^{90}Sr in the urine sample (the dashed lines indicate the spiked level with 95% CI).

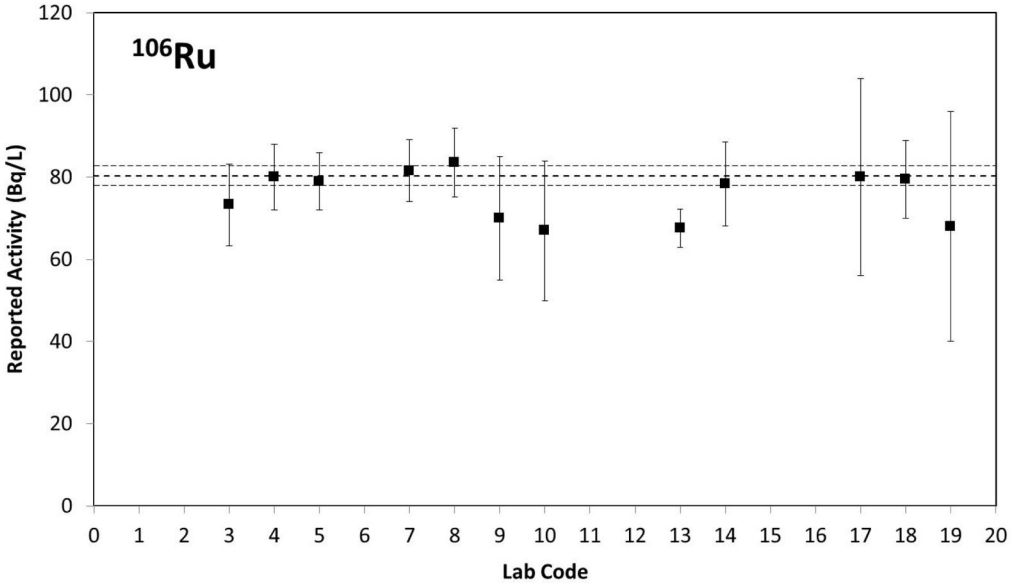

Figure 2.
Distribution of the reported results for ^{106}Ru in the urine sample (the dashed lines indicate the spiked level with 95% CI).

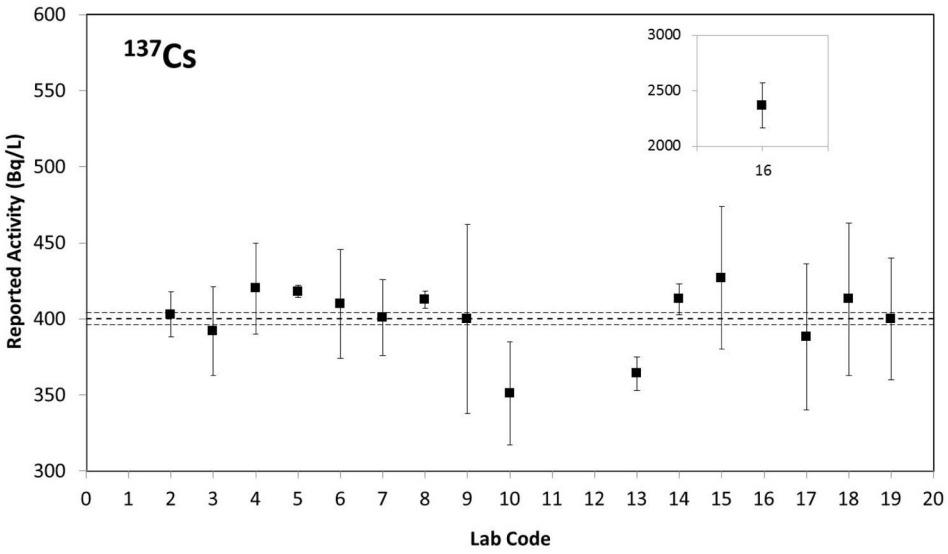

Figure 3.

Distribution of the reported results for ^{137}Cs in the urine sample (the dashed lines indicate the spiked level with 95% CI). Note that for L16, as the reported result is very different from the others, it is presented here as an insert.

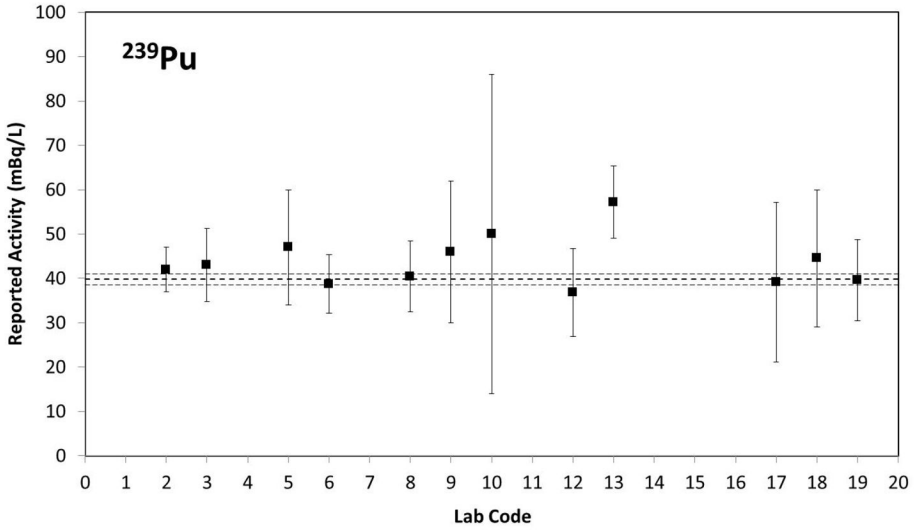

Figure 4.
Distribution of the reported results for ^{239}Pu in the urine sample (the dashed lines indicate the spiked level with 95% CI).

Table 1

Spiked activities (Bq) of the four radionuclides in a 250 mL sample provided to each participating laboratory and their concentration (Bq/L) (95% CI)

Radionuclide	$^{90}Sr^{*}$	$^{106}Ru^{*}$	^{137}Cs	^{239}Pu
Activity in a 250 mL sample (Bq)	5.02 ± 0.03	20.1 ± 0.6	100 ± 1	0.00994 ± 0.00003
Concentration (Bq/L)	20.08 ± 0.12	80.4 ± 2.4	400 ± 4	0.03976 ± 0.00012

*$^{90}Sr/^{90}Y$ and $^{106}Ru/^{106}Rh$ are in equilibrium

Table 2

Reported bioassay results (95% CI) from the participating laboratories

Lab Code	^{90}Sr (Bq/L)	^{106}Ru (Bq/L)	^{137}Cs (Bq/L)	^{239}Pu (mBq/L)
L01				<LoD *
L02			403 ± 15	42 ± 5
L03		73.3 ± 9.9	392 ± 29	43 ± 8.2
L04	17 ± 1.3	80 ± 8	420 ± 30	
L05	20 ± 2	79 ± 7	418 ± 4	47 ± 13
L06	19.6 ± 2.7		410 ± 36	38.7 ± 6.6
L07		81.6 ± 7.6	401 ± 25	
L08		83.6 ± 8.4	413 ± 5.6	40.4 ± 8
L09	50 ± 25	70 ± 15	400 ± 62	46 ± 16
L10	19.8 ± 3.3	67 ± 17	351 ± 34	50 ± 36 **
L12	17.9 ± 1.3			37 ± 10
L13	20 ± 2	67.6 ± 4.7	364 ± 11	57.2 ± 8.1
L14		78.4 ± 10.2	423 ± 10	
L15			427 ± 47	
L16			2368 ± 206	
L17	7.2 ± 1.6	80 ± 24	388 ± 48	39.2 ± 18
L18		79.5 ± 9.5	413 ± 50	44 ± 15
L19	21.6 ± 13.6	68 ± 28	400 ± 40	39.6 ± 9.2

* LoD was reported as 70 mBq/L;

** LoD was reported as 30 mBq/L