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Determination of ^{241}Am in Urine Using Sector Field Inductively Coupled Plasma Mass Spectrometry (SF-ICP-MS)

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Abstract

Quantification of ^{241}Am in urine at low levels is important for assessment of individuals' or populations' accidental, environmental, or terrorism-related internal contamination, but no convenient, precise method has been established to rapidly determine these low levels. Here we report a new analytical method to measure ^{241}Am as developed and validated at the Centers for Disease Control and Prevention (CDC) by means of the selective retention of Am from urine directly on DGA resin, followed by SF-ICP-MS detection. The method provides rapid results with a Limit of Detection (LOD) of 0.22 pg/L (0.028 Bq/L), which is lower than 1/3 of the C/P CDG for ^{241}Am at 5 days post-exposure. The results obtained by this method closely agree with CDC values as measured by Liquid Scintillation Counting, and with National Institute of Standards Technology (NIST) Certified Reference Materials (CRM) target values.

Introduction

Americium is a man-made, radioactive, metallic element produced when plutonium atoms undergo successive neutron capture events in nuclear reactors, in nuclear weapons, and during nuclear weapons' detonations. Americium has several different isotopes, all of which are radioactive. The most important and prevalent americium isotope is ^{241}Am , with a half-life of 432.7 years. As it decays, ^{241}Am releases alpha particles at 5.44 MeV (13%) and 5.49 MeV (84.5%), becoming ^{237}Np , which (in 35.9% of the decays) immediately emits gamma radiation at 59.5 keV. The ^{241}Am decay chain ends with ^{209}Bi , a nonradioactive element. ^{241}Am in the environment originated from atmospheric testing of nuclear weapons during the 1950s and 1960s, as well as reprocessing plants and nuclear accidents. Facilities that are involved with nuclear weapons, well logging sources and manufacture smoke detectors are minor sources of ^{241}Am contamination [1].

The critical ^{241}Am exposure pathways are inhalation and ingestion. ^{241}Am poses significant health hazards, even in small concentrations, if it is taken into the body in a soluble form. Once in the body, ^{241}Am concentrates in the skeleton, liver, and muscle. It can stay in the body for decades and continue to expose the surrounding tissues to both alpha and gamma radiation. Long-term internal exposure to ^{241}Am may create an increased risk of developing

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cancer. Exposure to any significant amount of ^{241}Am is unlikely under normal circumstances [1, 2].

Several techniques exist for the determination of ^{241}Am concentration in environmental and human samples [3, 4]. Gamma spectrometry, using High Purity Germanium detectors, is the primary tool used to determine ^{241}Am at levels of 0.1–1 Bq/kg or higher; but to obtain accurate results, it requires that the user correct for the attenuation of gamma rays in the samples [5, 6]. Alpha spectrometry is the most commonly applied technique for determination of low-level ^{241}Am content. Its principal advantages are relatively low equipment costs, high sensitivity due to low background, and high selectivity for alpha particles against other types of radiation [3]. A limit of detection (LOD) of 10–20 mBq/kg has been reported for various sample matrices, depending on the counting time and count rate of the procedure blank [7–9]. However, tedious, time-consuming sample preparation procedures (e.g., precipitation, evaporation, elution, filtration, electroplating, etc.) and long measurement times limit throughput. Such preparation procedures are due to possible interference from other radionuclides with close alpha energies, and long counting times are necessary because of ^{241}Am 's relatively low specific activity.

SF-ICP-MS offers substantial advantages over conventional radiometric techniques and has recently been used for analysis of many long-lived radionuclides in various sample matrices. It is one of the fastest methods for ^{241}Am analysis [10–13]. An LOD of 1 pg/L for ^{241}Am has been reported on SF-ICP-MS [14]. This LOD is comparable to that of alpha spectrometry, assuming no interferences exist for SF-ICP-MS. However, the main analytical issue in SF-ICP-MS originates from isobaric and polyatomic interferences such as $^{241}\text{Pu}^+$, $^{240}\text{PuH}^+$, $^{209}\text{Bi}^{32}\text{S}^+$, $^{209}\text{BiO}_2^+$, $^{206}\text{Pb}^{35}\text{Cl}^+$, $^{204}\text{Pb}^{37}\text{Cl}^+$, $^{205}\text{Tl}^{36}\text{Ar}^+$, $^{207}\text{Pb}^{34}\text{S}^+$, and $^{201}\text{Hg}^{40}\text{Ar}^+$ [11]. The major isobaric interference with ^{241}Am is ^{241}Pu . Since ^{241}Am is the decay product of ^{241}Pu (half-life is 14.33 years), in some samples of reactor origin the concentration of ^{241}Am is comparable to that of ^{241}Pu . Therefore, the method includes a thorough chemical separation of ^{241}Pu (which also removes most of the other interfering molecular ions) from the samples [11, 15].

Developing methods to determine exposure to ^{241}Am is within CDC's public health mission. Quantitative analysis of ^{241}Am in urine is considered a useful, noninvasive way to assess levels of internal contamination. Our Emergency Response Analytical goal is to be able to detect threat-radionuclides in urine at levels well below (i.e., 1/3 of or lower) the levels for a general population or for special subgroups such as children or pregnant woman (C/P) at the National Council on Radiation Protection & Measurements (NCRP) Report No. 161 Clinical Decision Guide (CDG) based action level of 0.73 pg/L (0.093 Bq/L) for ^{241}Am (urine output expected at 5 days post intake) [16]. CDC's IRATB recently developed a urine Gross Alpha/Gross Beta method using Liquid Scintillation Counting (LSC) [17]. However, the LOD of this method for ^{241}Am is 4.2 Bq/L, equivalent to 32.3 pg/L, which is much higher than the C/P CDG of 0.73 pg/L for ^{241}Am .

In this study, we report a novel and rapid analytical method for determination of ^{241}Am in urine samples. The Solid Phase Extraction (SPE) part of the method is based on preliminary studies carried out by Horwitz, et al. [18], Li, et al. [19] and Sadi, et al. [20] using a single

DGA resin cartridge to separate Am from other actinides such as U and Pu. We further optimized the method to isolate ^{241}Am from a 10-mL volume of urine using simple extraction steps and used SF-ICP-MS for detection instead of LSC. The study's purpose was to develop a rapid, simple method to address and respond to public health or other accidental, environmental, or terrorism-related exposures to ^{241}Am . This method is not designed to characterize the normal background level of ^{241}Am in the non-occupationally exposed population, but it does have a detection limit below the suggested CDG action levels. Thus it can serve as a means of rapidly identifying both adults and children who have been exposed to ^{241}Am and who might require medical intervention.

Experimental

Reagents and solutions

DGA Cartridges (normal, 1 mL) and a polycarbonate vacuum box (24 holes) were purchased from Eichrom Technologies (Darien, IL, USA). All nitric (HNO_3) and hydrochloric (HCl) acid solutions were prepared from double-distilled (DD) acids (GFS Chemicals Inc. Columbus, OH). Deionized water was used for all solutions ($< 18 \text{ M}\Omega\text{-cm}$, from an Aqua Solutions Ultrapure Water System, Aqua Solutions, Inc., Jasper, GA). "Base urine" was collected through anonymous human donations (CDC protocol 3994) and acidified to 1% v/v HNO_3 . All radioactivity solution sources were traceable to the National Institute for Standards and Technology (NIST, Gaithersburg, MD, USA). Both low and high quality control (QC) solutions and other urine pools for LOD were prepared for determination by spiking base urine with dilutions of an ^{241}Am isotope standard (Eckert & Ziegler Analytics, Inc., Atlanta, GA). A series of aqueous ^{241}Am Certified Reference Materials (CRM) solutions were prepared by dilution of ^{241}Am radioactive source solutions from NIST. ^{243}Am (Eckert & Ziegler Analytics Inc., Atlanta, GA) was used as an internal standard (tracer). Sodium nitrite (Sigma-Aldrich, St. Louis, MO) was used to adjust the oxidation states. Serial dilutions of uranium, lead, thallium, mercury, bismuth single-element stock standards (SPEX Industries, Inc., Edison, NJ) and a ^{242}Pu radioactivity solution (U.S. Department of Energy, New Brunswick Laboratory, Argonne, IL) were spiked into the urine samples to verify that high separation factors for U, Pb, Tl, Hg, Bi and Pu were obtained using this SPE procedure.

Sample preparation

The urine sample volume for a single analysis is 10 mL. Allow urine specimens to reach ambient temperature, shake or vortex them to mix for 5 seconds before pipetting. Spike 400 μL of 1 ng/L ^{243}Am solution as an internal standard (tracer) to every 10 mL of urine patient sample or QC sample. Add 4.76 mL of concentrated HNO_3 (68–70%, the final concentration in the sample is 5M) and then 0.13g of sodium nitrite to each sample as a valence adjuster to convert Pu to the tetravalent state. Shake or vortex to mix the samples for 5 seconds and let reaction occur at room temperature for at least 10 minutes. Load each sample on a DGA resin cartridge of 1 mL bed volume (cartridge preconditioned with 15 mL of 5 M HNO_3 using a vacuum box). Rinse the cartridge again with 15 mL of 5M HNO_3 followed by 15 mL $\times 3$ of 0.5M HNO_3 using a vacuum box. Strip ^{241}Am from the column with 5 mL of 0.5M HCl . Transfer 1 mL of the purified samples into 4 mL polystyrene conical bottom sample

cups for analysis (Figure 1). Prepare external, aqueous-based stock calibration standards by spiking 0.5M HCl with dilutions of ^{241}Am isotope standard, and then add 40 μL of internal standard solution (1 ng/L ^{243}Am) to every 1 mL of standards to reach the same tracer concentration as the patient and QC samples. Prepare both calibration standards and sample blanks as 0.5 M HCl solutions, which match the elute solutions for the column of this method.

Instrumentation

This method measures ^{241}Am concentrations using an extended dynamic range, high-resolution ICP-MS model Element XR (Thermo Fisher Scientific, Bremen, Germany), which is a double-focusing, magnetic sector, inductively-coupled-plasma mass spectrometer with a single discrete dynode detector (Mascom, Bremen, Germany). It uses the ICP-MS, equipped with nickel sampler and skimmer cones and a CD-2 guard electrode, in triple mode. The sample introduction system consists of a computer-controlled ASX-112 (Cetac, Omaha, NE) autosampler and an Aridus IITM (Cetac, Omaha, NE) desolvation unit. As discussed in our lab's previous report [21], the Aridus IITM setup increases the sensitivity of the SF-ICP-MS by more than 10 times, enabling the measurement of ^{241}Am at the low level of < 1 pg/L. Samples self-aspirate from the autosampler into the desolvation unit through an Apex perfluoroalkoxy (PFA) 100 $\mu\text{L}/\text{minute}$ nebulizer (ESI, Omaha, NE, or equivalent). The desolvation unit, equipped with an upgraded PFA spray chamber, operates at 110 $^{\circ}\text{C}$. With the aid of argon sweep gas and nitrogen gas for sensitivity enhancement, the sample passes through a semi-permeable membrane coil in the unit that operates at 160 $^{\circ}\text{C}$. Optimize flow rates as needed, with argon sweep gas at $\sim 3\text{--}7$ L/min and nitrogen gas at $\sim 3\text{--}7$ mL/min. The desolvated sample exits the unit into a 1.8 mm I.D. sapphire injector and a standard quartz torch, and then into the mass spectrometer. All experimental parameters are optimized for ^{241}Am concentrations determination by SF-ICP-MS with respect to maximum ion intensity of ^{238}U and minimum uranium oxide formation rate using a 5 ng/L natural uranium tuning solution. Table 1 contains a summary of our optimized operating conditions.

Results and discussion

Removal of potential spectral interferences

Potential interferences for analysis of ^{241}Am include isobaric overlaps with anthropogenic ^{241}Pu and polyatomic overlaps with $^{240}\text{PuH}^{+}$, $^{209}\text{Bi}^{32}\text{S}^{+}$, $^{209}\text{BiO}_2^{+}$, $^{206}\text{Pb}^{35}\text{Cl}^{+}$, $^{204}\text{Pb}^{37}\text{Cl}^{+}$, $^{205}\text{Tl}^{36}\text{Ar}^{+}$, $^{207}\text{Pb}^{34}\text{S}^{+}$, and $^{201}\text{Hg}^{40}\text{Ar}^{+}$. To test for complete removal of ^{241}Pu , a 50 pg/L solution of ^{242}Pu isotope spike in base urine was prepared and tested. Experiments showed more than 99% of ^{242}Pu is removed by the SPE portion of sample preparation. Using SPE sample preparation as described above, Pb, Tl, and Hg, spiked in base urine at concentrations of 3 $\mu\text{g}/\text{L}$, 0.5 $\mu\text{g}/\text{L}$, and 5 $\mu\text{g}/\text{L}$ respectively, did not result in apparent (> 0.1 pg/L) ^{241}Am concentrations. These spiked urine samples' concentrations were above the National Health and Nutrition Examination Survey (NHANES) 95th percentile of urine Pb, Tl, and Hg concentrations [22]. Although no NHANES survey data was available for bismuth, analysis of what was otherwise determined [23, 24] to be a high urine concentration (5 $\mu\text{g}/\text{L}$) of Bi, produced no apparent ^{241}Am concentration.

Performance of a natural U-spike experiment determined that due to peak tailing, small interferences remained at $m/z = 241$ when the separated sample solutions contain high levels of U ($> 0.5 \mu\text{g/L}$). Analysis of urine samples with U $= 1.0 \mu\text{g/L}$ with this SPE method as part of the sample preparation procedure removed more than 99% of the U that might cause tailing into the $m/z=241$ region. Samples having U concentrations higher than $10.0 \mu\text{g/L}$ (the NHANES 95th percentile of U concentration in urine of normal U. S. residents is $0.031 \mu\text{g/L}$) [22] should be treated by the modified sample preparation procedure as shown in Figure 1, which will be described in more detail below.

Limit of detection

The LOD for ^{241}Am in urine specimens is based on 60 analytical runs of 4 different low-concentration samples close to the LOD (a first approximation of LOD is the measured blank concentration plus 3 times the Standard Deviation (SD) of the measured blank concentration) and was calculated according to the formula:

$\text{Conc}_{\text{LOD}} = [\text{mean}_b + 1.645(\text{Sb} + \text{int})]/[1 - 1.645(\text{slope})]$, where mean_b = blank average, Sb = standard deviation of blank average, int = intercept of the equation of SD versus concentration for LOD samples analyzed at least 60 times, Slope = slope of the equation of SD versus concentration for LOD samples analyzed at least 60 times.

The LOD of this method is 0.22 pg/L (Figure 2). This LOD is $< 1/3$ of the C/P CDG ($\sim 0.734 \text{ pg/L}$), and is therefore acceptable for an emergency radiobioassay method for determining the concentration of ^{241}Am in urine collected at 5 days post-exposure.

Linearity

A linearity study determined the linear reportable range for this method. The method exhibits good linear signal response between concentrations of 0.3 pg/L and 1000 pg/L of ^{241}Am with a Coefficient of Determination of 1.000. The normal calibration range is from 0.3 pg/L to 30 pg/L , and the extended calibration range is from 30 pg/L to 1000 pg/L . If a urine ^{241}Am value is above the highest calibrator, the urine sample is diluted with 5% HNO_3 to bring the concentration within the validated calibration range.

Internal methods comparison study

A comparison of urine sample analysis results was performed between this method and our CLIA validated LSC method. The two samples LU-077203 and HU-077201 were prepared as QC material and, using LSC, analyzed for ^{241}Am at relatively high concentrations. They then were diluted 1:1000 to get within the desired ^{241}Am concentration range for the present method, purified and analyzed using SF-ICP-MS. The difference between the described methods is 2.1% to 3.0% (Table 2),

Precision and accuracy

Analysis of serial aqueous dilutions of a Certified Reference Material (CRM) from NIST was also used to verify method accuracy. The observed ^{241}Am concentrations were in close agreement with the target values, with an analytical bias from -0.3% to 1.7% (Table 2). Table 2 also shows the typical precision observed at different concentrations of daily quality

control materials analyzed at the beginning, in the middle, and at the end of each analytical run. Accuracy and precision of the reported results was assured based on adherence to the quality control/quality assurance program of the Division of Laboratory Sciences, NCEH, CDC [25].

Analysis of samples from the NIST Radiochemistry Intercomparison Program (NRIP)

NRIP is a performance evaluation program which provides high quality, traceable radionuclide materials to support low-level radioanalytical laboratories conducting environmental and radiobioassay radioactivity measurements. ^{241}Am is among the radionuclides used for testing. However, we found that the extraordinarily high concentrations of uranium present in these samples (intended for evaluation of environmental levels of uranium by alpha spectrometry) significantly affects the accuracy of trace level ^{241}Am determination by SF-ICP-MS. Further, these uranium concentrations would possibly produce significant, troublesome instrument contamination. To address this problem we developed and recommend a modified sample preparation procedure that is further optimized for samples with extremely high U content (usually higher than 10 $\mu\text{g/L}$).

In this procedure, after rinsing the cartridge with 15 mL of 5M HNO_3 followed by 15 mL \times 3 of 0.5M HNO_3 , replace both the cartridge reservoirs and tips to eliminate possible U deposits and rinse the cartridges with more 0.5M HNO_3 (15 mL \times (3 - 6) of 0.5M HNO_3 , see Figure 1). Table 3 and Table 4 show the results observed for ^{241}Am analysis of the NRIP samples. These samples were from two radiobioassay preparedness exercises during 2012 with different turnaround times (TATs): one 60 days, and one 8 hours. These synthetic urine samples typically have U concentrations ranging from 140 $\mu\text{g/L}$ to 450 $\mu\text{g/L}$. After the more aggressive rinsing procedure, U concentrations in the elution solutions were under 0.20 $\mu\text{g/L}$, and did not result in apparent ^{241}Am signal contribution for these samples. All but one result had slight negative bias (average $-2.1 \pm 2.4\%$ at a 95% Confidence Level) compared with the NIST target values. Most of the observed results for the NRIP samples show a small negative bias compared to the NIST target values, indicating a slight negative systematic uncertainty. One result had a positive bias of 12.7%. We noted that analyses of this sample for other radionuclides yielded a similar positive bias, indicating an external sample preparation error, as opposed to method bias.

Sample turnaround time (TAT)

While maintaining high quality results, sample TAT is one of the important considerations in a radiological emergency. For this method, ~ 2.5 hours are required to pretreat the urine samples for a batch of 20 patient urine specimens plus QC samples. An additional 3.5 hours are required for final analysis of 20 patient samples by SF-ICP-MS, including calibrators, blanks, and QC samples. Samples may be pretreated concurrently with final SF-ICP-MS analysis, resulting in a daily throughput of approximately 120 samples per day (24 hours) per instrument.

Conclusions

We introduced a method for rapidly determining ultra-low levels of ^{241}Am in urine samples using a Solid Phase Extraction purification procedure and a high-sensitivity sample introduction system (Aridus IITM), coupled with SF-ICP-MS. This method provides for analysis of ^{241}Am at very low levels, with a LOD of 0.22 pg/L (well below the C/P CDG level) and allows rapid throughput of samples. It attained good agreement, with a bias of 2.1%–3.0%, for urine samples in an internal comparison with a CDC LSC method. It also produced recoveries from 99.7% to 101.7% in analysis of aqueous dilutions of ^{241}Am SRM from NIST.

This method's efficient urine sample separation scheme effectively eliminates most molecular ion interferences. However, if urine samples contain more than 10 $\mu\text{g/L}$ of U, more aggressive rinsing procedures are required to eliminate U from the elution solutions. The results obtained by this method for NIST/NRIP reference materials with high-U levels are in close agreement with the NIST target values, with biases ranging from –0.62% to –5.61%.

A major advantage of this method over alpha spectrometry and other methods is that only a small, 10 mL volume of each urine sample is needed to perform the analysis, making successful analysis more likely, especially for young children and infants.

This procedure is appropriate for rapid identification and quantification of ^{241}Am in urine for emergency response involving accidental or terrorism-related elevated exposures, or for evaluating chronic environmental or other non-occupational exposures.

Acknowledgments

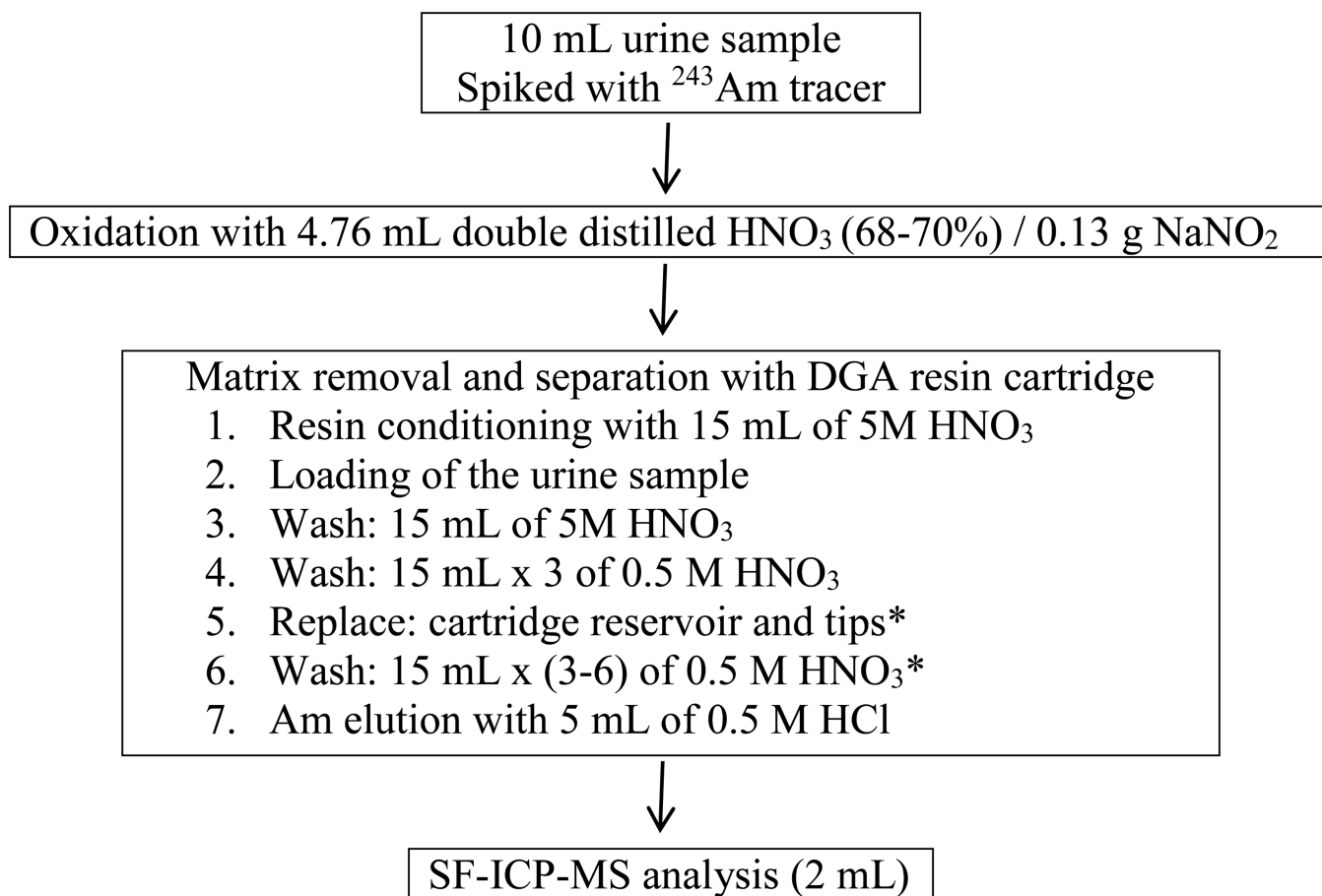
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**Figure 1.**Sequential sample preparation procedure for ^{241}Am determination* Samples containing U concentrations greater than 10 $\mu\text{g/L}$, add steps 5 – 6.

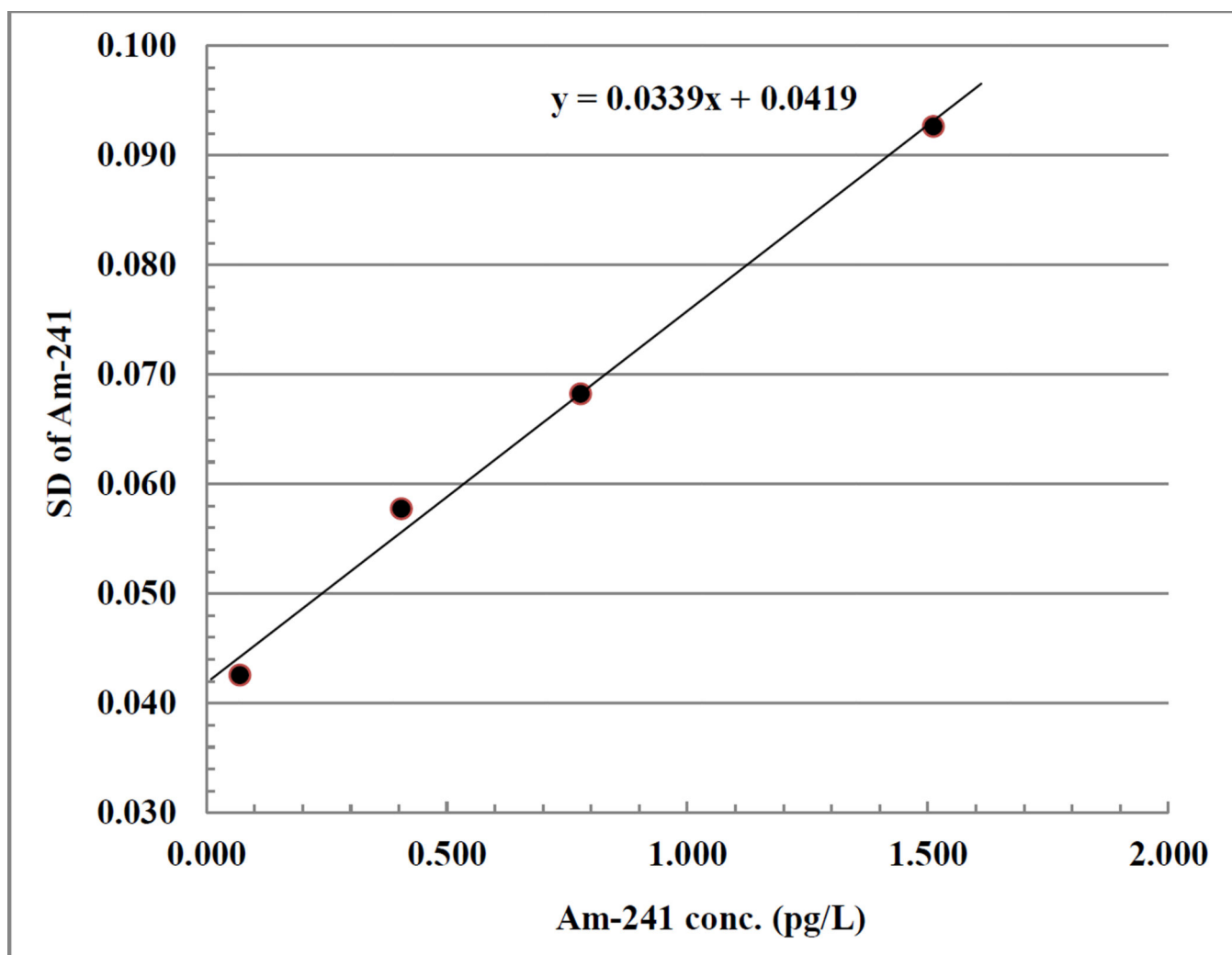


Figure 2.
Plot for ^{241}Am LOD determination (60 runs per point).

Table 1

Instrumental conditions and data acquisition settings for SF-ICP-MS measurements

RF Power (KW)	1.2 – 1.3
Cooling Gas flow (L/min)	16
Auxiliary Gas flow (L/min)	0.9
Sample Gas flow (L/min)	0.7 – 0.8
Lenses (V)	Optimized as needed
Sample Take up time (min)	2.1
Wash (min)	3
Pump Speed During Wash (rpm)	1
LR Runs/Passes	3* 60
Detection Mode	Triple
Measurement Units	CPS
Scan Type	ESCAN
Scan Optimization	Speed
Number of Pre-Scans	5
Integration Type	Average
Res. Switch Delay (s)	2
Resolution	300
Mass Window (%)	15
Setting Time (s)	0.001
Sample Time (s)	0.001
Samples Per Peak	200
Search Window (%)	20
Integration Window (%)	15
Measured Isotopes	²⁴¹ Am, ²⁴³ Am

Table 2

Observed ²⁴¹Am concentrations (pg/L) and among-run precision for reference materials and internal quality control materials

Sample	²⁴¹ Am			
	N	Average	SD	Target Value Bias (%)
LU-077203 ^a	12	8,220	310	8,050 ^d 2.1
HU-077201 ^a	12	20,700	600	20,100 ^d 3.0
Pool1 ^b	59	0.335	0.058	0.3 ^e 12
Low QC ^c	120	0.708	0.068	0.7 ^e 1.2
Pool2 ^b	60	1.44	0.093	1.4 ^e 3.0
High QC ^c	120	10.1	0.56	10.0 ^e 0.9
Extended High QC ^c	40	783	33.6	800 ^e -2.1
Dilution 1 ^f	1	100	-	100 0.0
Dilution 2 ^f	1	203	-	200 1.7
Dilution 3 ^f	1	300	-	300 -0.1
Dilution 4 ^f	1	399	-	400 -0.3
Dilution 5 ^f	1	605	-	600 0.8
Dilution 6 ^f	1	1007	-	1000 0.7

^a 1:1000 dilution of urine QC materials used for the LSC Gross Alpha/Beta method at CDC.

^b Urine materials made at CDC by spiking certified reference material in pooled urine collected anonymously.

^c Internal quality control materials made at CDC by spiking certified reference material in pooled urine collected anonymously.

^d Characterized results of co-worker by using the LSC Gross Alpha/Beta method at CDC[17].

^e Target values of spiked urine pools using certified reference material.

^f Aqueous dilutions of CRM from NIST.

Table 3Comparison of CDC ^{241}Am results with NIST target values for NRIP12 60 days samples ^{*}

Sample ID	Massic Activity (NIST Target Value)	Massic Activity (CDC Observed Results)	Relative Expanded Uncertainty (k=2)	Bias
	(Bq/g spike)	(Bq/g spike)	(%)	(%)
207	1.784	1.74	11.5	-2.52
212	1.784	1.74	11.2	-2.41
220	1.784	1.76	12.8	-1.57
224	1.784	1.74	12.5	-2.52
227	1.784	1.76	12.1	-1.63
214	1.784	1.74	12.3	-2.47
216	1.784	1.71	12.1	-4.09
228	1.784	1.75	12.2	-2.02
231	1.784	1.77	11.5	-0.95
232	1.784	1.77	11.2	-0.73
208	1.784	1.74	13.1	-2.75
211	1.784	1.74	13.0	-2.58
219	1.784	1.68	13.8	-5.61
223	1.784	1.76	11.7	-1.57
226	1.784	1.77	11.8	-1.01

* All samples were diluted 1:2 before DGA (Eichrom's extraction chromatographic materials in which the extractant system is N,N,N',N'-tetra-n-octyldiglycolamide resin) separation

Table 4Comparison of CDC ^{241}Am results with NIST target values for NRIP12 8 hours samples *

Sample ID	Massic Activity (NIST Target Values)	Massic Activity (CDC Observed Results)	Relative Expanded Uncertainty (k=2)	Bias
	(Bq/sample)	(Bq/sample)	(%)	(%)
215	0.146	0.14	11.0	-1.93
218	0.292	0.29	10.9	-0.62
222	0.149	0.15	11.1	-1.47
230	0.297	0.29	11.1	-1.99
234 **	0.372	0.42	11.2	12.7

* Samples 218 and 230 were diluted 1:2 and sample 234 was diluted 1:4 before DGA (Eichrom's extraction chromatographic materials in which the extractant system is N,N,N',N'-tetra-n-octyldiglycolamide resin) separation

** Analyses for other radionuclides also produced unusually high results for this sample.