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International Journal of Mass Spectrometry 178 (1998) 113–120

International Journal of Mass Spectrometry

Applications of thermal ionization mass spectrometry to the detection of ^{239}Pu and ^{240}Pu intakes

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Received 20 May 1998; accepted 18 June 1998

Abstract

The United States Department of Energy requires routine bioassay monitoring for workers who may incur intakes of radioactive materials, that may result in a committed effective dose equivalent of 1 mSv. The radiochemistry/α-spectroscopy method, historically used for analysis of plutonium in urine, does not provide the level of measurement sensitivity to meet this monitoring requirement. Los Alamos National Laboratory has an established, ultrasensitive, actinide analysis program. Application of class-100 clean room radiochemistry and thermal ionization mass spectrometry to the determination of plutonium concentration in human urine samples yielded an average measurement uncertainty of 3.8 μBq 24 h^{-1}, a 40-fold improvement over the measurement uncertainties associated with radiochemistry/α-spectroscopy analytical methods. This measurement capability corresponds to an ability to detect intakes on the order of 30 Bq of ^{239}Pu and ^{240}Pu, under conditions of annual routine monitoring. The resulting minimum detectable committed effective dose equivalent associated with this intake is 2 mSv. (Int J Mass Spectrom 178 (1998) 113–120) © 1998 Elsevier Science B.V.

Keywords: Plutonium bioassay; Plutonium intake estimation; Plutonium; Mass spectrometry

1. Introduction

Current United States Department of Energy (DOE) regulations require routine bioassay monitoring for all workers who have a reasonable potential for intakes of radioactive materials in a single year that may result in a committed effective dose equivalent (CEDE) of 1 mSv. The term “monitoring” is clearly defined in the Code of Federal Regulations as “the measurement of quantities . . . of radioactive material and the use of the results . . . to evaluate potential and actual exposures” [1].

The historically accepted method for detecting occupational intakes and assessing the dosimetric consequences of plutonium inhalation is by radiochemical evaluation of urine samples collected from the individual [2–10]. Detection of α-particle emissions from the decay of plutonium in chemically prepared urine samples has been the technique of choice since the early days of the Manhattan District [11]. Alpha-spectroscopy analysis of urine samples has a measurement uncertainty on the order of 150 μBq day^{-1} and an associated CEDE in the range of 20 to 100 mSv, for routine biannual sampling in the year of intake [9,10,12,13].

Relative to currently accepted radiation dose lim-

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PII S1387-3806(98)14084-8

its, detection of ^{239}Pu by in vivo lung counting is limited by a measurement uncertainty on the order of 400 Bq for ^{239}Pu. This uncertainty translates into an intake of approximately 15 kBq and a CEDE of greater than 1 Sv (if the measurement is made at 180 days after an acute inhalation intake of 1 μm activity median aerodynamic diameter (AMAD), inhalation class *Y*, ^{239}Pu) [10,14]. Detection by analysis of fecal samples has shown some theoretical promise; however, for practical applications, the associated biological variability and contributions from ingested plutonium produce intake estimates with unacceptably large statistical uncertainties [15,16].

In most cases, inhalation of ^{239}Pu, from environmental sources, occupational exposures occurring decades earlier, and low-level occupational exposures are not detectable by using radiochemical techniques [9]. In addressing this technical shortfall, the DOE has prescribed the use of "the best practical state-of-the-art bioassay monitoring methods" [17].

The LANL Nuclear and Radiochemistry group and the Environmental Systems and Waste Characterization group have a well-established ultrasensitive actinide analysis program. In addition to classical radiochemical techniques, this program utilizes class-100 clean room technology, ultratrace actinide chemistry, and thermal ionization mass spectrometry (TIMS) to detect ultralow levels of ^{239}Pu, ^{240}Pu, and other actinides in biological and environmental matrices [18–21].

The thermal ionization mass spectrometry (TIMS) measurement uncertainty for detecting ^{239}Pu and ^{240}Pu in biological samples is on the order of 1 μBq/sample ($\approx 10^6$ atoms of ^{239}Pu) [20]. A fission track method (FTM) for urine bioassay developed at the University of Utah and Brookhaven National Laboratory (BNL) has a reported measurement uncertainty of approximately 0.6 μBq/sample for fissionable materials in urine [9,22].

2. Methods

Human urine excreta were collected for a 24 h interval, from a population of individuals who work at Los Alamos National Laboratory (LANL). Also, synthetic urine samples traced with ^{239}Pu (3.7, 9.3, 29.6, and 55.5 μBq/sample) were received from Yankee Atomic [23,24].

Each 24 h urine sample was traced with ultrapure ^{242}Pu [25]. The plutonium fraction was coprecipitated with alkaline earth phosphate at room temperature. The precipitate was dissolved in 8 M HNO_3, heated, and adsorbed onto anion exchange resin. The plutonium was eluted from the anion exchange column with successive rinses of 0.5 M HCl, HI–HCl reagent and H_2O. The elute was then evaporated to dryness. The plutonium was dissolved in 12 M HCl containing a drop of H_2O_2 and adsorbed onto a second anion exchange column. The second column was rinsed with the HI–HCl reagent. The plutonium was then electroplated onto a platinum disk. The platinum disk was analyzed, under vacuum, with a 300 mm^2 solid state surface barrier detector for 6×10^5 s. The plutonium was removed from the platinum disk with HF and HNO_3. The sample was evaporated to dryness. The plutonium was dissolved in 12 M HCl containing a drop of H_2O_2 and adsorbed onto a third anion exchange column. The sample was rinsed with 8 M HNO_3 and the plutonium was eluted with a rinse of HBr. The sample was electrodeposited with platinum onto a rhenium ribbon filament and analyzed for ^{239}Pu and ^{240}Pu atom contents by thermal ionization mass spectrometry. All chemical processing was performed in class-100 clean rooms [26].

The synthetic urine samples were traced with ultrapure ^{242}Pu [25]. The plutonium fraction was coprecipitated with alkaline earth phosphate at room temperature. The precipitate was dissolved in 8 M HNO_3, heated, and adsorbed onto anion exchange resin. The column was rinsed with 8 M HNO_3. Plutonium was eluted from the anion exchange column with a 0.36 M HCl − 0.01 M F^- solution. The solution was evaporated to dryness. The plutonium was redissolved and then electroplated onto a platinum disk. The platinum disk was analyzed, under vacuum, with a 300 mm^2 solid state surface barrier detector for 7×10^4 s. The plutonium was removed from the platinum disk with HF and HNO_3. The sample was evaporated to dryness. The plutonium was dissolved in 12 M HCl containing a drop of H_2O_2

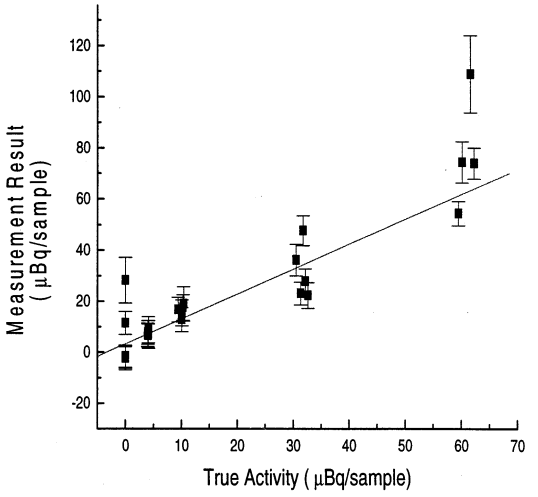

Fig. 1 Measured ^{239}Pu activity as a function of true sample activity. Results are from the TIMS analysis of synthetic urine samples. The line indicates a simple linear weighted fit of the data. The linear fit has an intercept of 3.3 ± 2.2 μBq, a slope of 0.98 ± 0.09, and a correlation coefficient of 0.92.

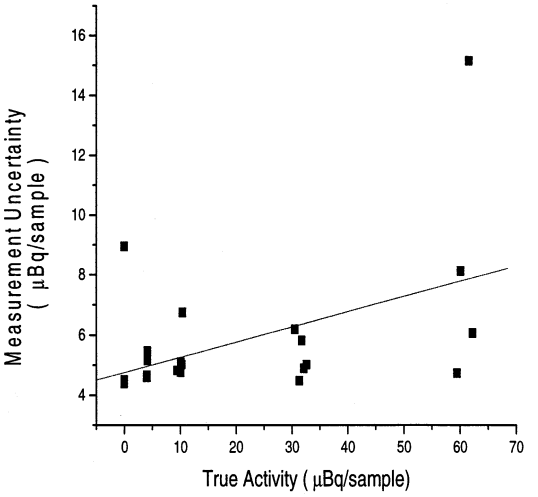

Fig. 2 Uncertainty of TIMS measured ^{239}Pu activity as a function of true sample activity. The line indicates a simple linear fit of the data. The linear fit has an intercept of 4.7 ± 0.6 μBq, a slope of 0.05 ± 0.02, and a correlation coefficient of 0.48.

and adsorbed onto a third anion exchange column. The sample was rinsed with 8 M HNO_3 and the plutonium was eluted with a rinse of HBr. The sample was electrodeposited with platinum onto a rhenium ribbon filament and analyzed for ^{239}Pu and ^{240}Pu atom contents by TIMS. All chemical processing was performed in class-100 clean rooms [26].

3. Results

3.1. Statistical evaluation of TIMS data

LANL participated in the DOE Office of International Health interlaboratory comparison for detecting ultralow levels of plutonium in human urine [24]. The α-spectroscopy analysis did not detect ^{239}Pu activity in any of the synthetic urine samples. Figure 1 contains a plot of the measured ^{239}Pu activity, by TIMS analysis, in each synthetic urine sample as a function of the known sample activity. Figure 2 contains a plot of the uncertainty of the measured ^{239}Pu activity, by TIMS analysis, as a function of the known sample activity. Detailed results are available in the NIST report [24].

Table 1 contains measured ^{239}Pu and ^{240}Pu concentrations for a sample of individuals at LANL. Individuals who work or have worked with ^{239}Pu and ^{240}Pu are indicated in Table 1. Table 1 also contains the collected and analyzed urine volumes. The average volume of urine collected over a 24 h period was 1801 ± 801 mL.

Figure 3 contains a plot of a convolution of a Gaussian measurement error distribution and a two component delta function-uniform prior distribution uniform in log space [27]. The delta function represents a nonoccupationally exposed population and the uniform distribution represents the occupationally exposed population [13,28]. The estimated TIMS measurement uncertainty based on this fit is 3.8 μBq 24 h^{-1}. Table 2 contains a comparison of the LANL TIMS and α-spectroscopy measurement capabilities for ^{239}Pu. The distributional analysis revealed an estimated measurement bias of 3.7 μBq 24 h^{-1} for the TIMS measurement process on human urine.

4. Discussion and conclusions

The current radiochemical α-spectroscopy technique used at LANL for plutonium urine bioassay has

Table 1
Summary TIMS urine assay results for a population of LANL workers. Results from this table are displayed in the histogram in Fig. 3. In cases where two samples were submitted by one individual the samples are denoted by the letters *a* and *b*. Urine samples were collected over 24 h period. Individuals who work with plutonium are indicated in the last column by the letter *Y*

ID	TIMS (μBq 24 h^{-1})	σ (μBq 24 h^{-1})	Analyzed volume (mL)	Collected volume (mL)	Plutonium work?
1	−0.7	0.4	1750	1990	*Y*
2	512.8	7.0	995	995	*Y*
3	67.0	4.4	1500	1835	*Y*
5	35.6	4.1	1865	1865	*Y*
9	−0.5	2.6	2395	2395	*Y*
10	−8.4	5.2	1750	3030	*Y*
14	3.0	3.0	1845	1845	*Y*
15	36.4	7.0	1500	3270	*Y*
17	9.1	3.0	1620	1620	*Y*
20	95.0	5.2	1750	2735	*Y*
21	62.0	3.7	1750	1995	*Y*
22	25.5	4.1	1730	1730	*Y*
23	47.4	5.2	1750	1985	*Y*
24	5.8	3.0	1530	1530	*Y*
26	6.8	3.0	535	535	*Y*
27	12.4	3.7	1142	1142	*Y*
31	−1.2	4.4	1750	2720	*Y*
32	105.8	3.3	680	680	*Y*
36	3.7	3.0	1065	1065	*Y*
37	79.0	4.8	1750	2590	*Y*
38	146.4	4.8	1070	1070	*Y*
40	1.6	4.1	1750	2135	*Y*
41	5.9	5.2	1500	2860	*Y*
42*a*	8.0	3.3	1340	1340	*Y*
42*b*	4.8	3.0	355	355	*Y*
43	19.2	5.6	1450	2920	*Y*
44	0.2	4.4	1750	2430	*Y*
45	115.1	5.6	1210	1210	*Y*
46	7.8	3.0	672	672	*Y*
47	439.9	7.0	1515	1515	*Y*
49	105.4	13.3	1710	1710	*Y*
51	47.3	2.6	2465	2465	*Y*
52	50.8	2.2	2358	2358	*Y*
53	48.4	3.7	925	925	*Y*
54	39.1	3.0	3120	3120	*Y*
56	6.6	2.2	1258	1258	*Y*
57	13.6	2.2	3155	3155	*Y*
58*a*	−0.6	2.2	987	987	*Y*
58*b*	10.3	2.6	910	910	*Y*
60	7.0	2.2	2330	2330	*Y*
61	19 424.6	158.0	1675	700	*Y*
62	2 563.0	41.1	1430	700	*Y*
63	41.8	35.2	2460	700	*N*
64	64.0	51.1	3270	700	*N*
65	300.8	13.7	775	700	*Y*
66	770.3	55.1	2715	700	*Y*
67	1.5	18.1	1340	700	*N*
68	−2.2	29.6	1950	700	*N*
69	20.4	22.9	1030	500	*N*
70	58.1	38.1	2235	700	*N*

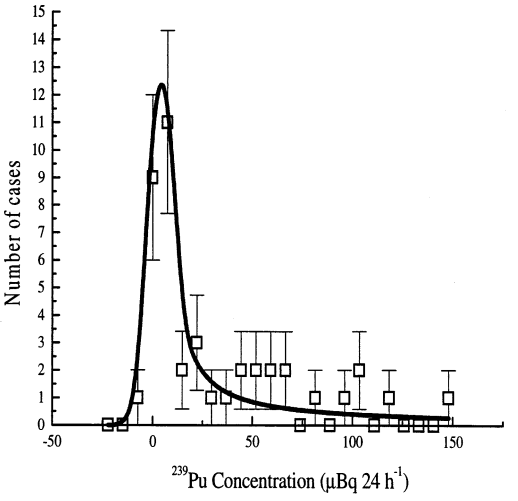

Fig. 3 Histogram of TIMS ^{239}Pu urine bioassay measurements from population of workers at LANL. The solid line represents a convolution of a Gaussian distribution representing measurement uncertainties and a delta-uniform prior distribution, (the uniform is in log space) representing the distribution of true amounts of plutonium in the samples. The error bars represent the binned data uncertainty as the square root of the number of events in a bin. The TIMS measurement error based on the fit is 3.8 μBq 24 h^{-1}.

a measurement uncertainty of 150 μBq per 24 h urine sample [13]. This measurement capability corresponds to detecting an intake of 1 kBq, and a CEDE of 100 mSv (if the measurement is made at 180 days after an acute inhalation intake of 1 μm AMAD, inhalation class *Y*, ^{239}Pu). Application of the class-100 clean room radiochemistry and thermal ionization mass spectrometry to the determination of plutonium concentration in the collected urine samples yielded an average measurement uncertainty of 3.8 μBq 24 h^{-1} and a measurement bias of 3.7 μBq 24 h^{-1}. This measurement capability corresponds to detecting an intake of 30 Bq for annual routine monitoring and a CEDE of 2 mSv.

Table 2
Summary of LANL bioassay techniques including the average measurement error, and the minimum detectable dose at 180 days after an acute intake of inhalation class *Y*, 1 μm AMAD ^{239}Pu, for a single annual urine sample

Method	σ_0 (μBq 24 h^{-1}) [atoms 24 h^{-1}]	CEDE (mSv)
TIMS Human urine	3.8 [4.2 × 10^6]	1.5
TIMS Synthetic urine	4.8 [5.3 × 10^6]	1.7
α-spec Human urine	150 [2 × 10^8]	55

In general, the results from the DOE Office of International Health interlaboratory comparison were promising and validated the use of the single stage TIMS instrument for monitoring plutonium workers. The estimated measurement uncertainty for the blank synthetic urine samples was 4.8 μBq ± 0.6 μBq. This result is comparable to the estimated synthetic urine measurement uncertainty of 3.8 μBq 24 h^{-1}, for human urine samples. The TIMS measurement bias for synthetic urine was taken as the *y* intercept in Fig. 1. This measurement bias of 3.3 μBq ± 2.2 μBq for synthetic urine is comparable to the estimated measurement bias of 3.7 μBq 24 h^{-1} in human urine.

To test the influence of a lower measurement error on the interpretation of bioassay samples, the TIMS, and α-spectroscopy techniques were compared by using distributions of random urine excretion patterns generated by a series of Monte Carlo experiments. In the first case, the assumed intake scenario was an acute inhalation of 12 Bq, 1 μm AMAD, inhalation class *Y*, ^{239}Pu. It was further assumed in our analysis that urine samples were collected on days 1, 3, and 5 after the intake for the TIMS analysis protocol. The samples from the TIMS protocol were generated with a measurement uncertainty of 3.8 μBq per 24 h urine sample. For the α-spectroscopy analysis, samples were collected on days 1, 2, 4, 8, 16, 30, 60, 120, and 240 after the intake. The samples from this second protocol were generated with a measurement uncertainty of 150 μBq per 24 h urine sample. A biological variation of 10% was used for both data sets. The ICRP Publication 30 lung model and the Jones plutonium urine excretion model were used in the Monte Carlo calculations to generate two sets of 100 random realizations of plutonium concentrations in urine due to the intake scenario [4,5,29].

The two sets of random realizations were analyzed with a mathematical unfolding technique developed at Los Alamos [13,16,28,30,31]. The results are dis-

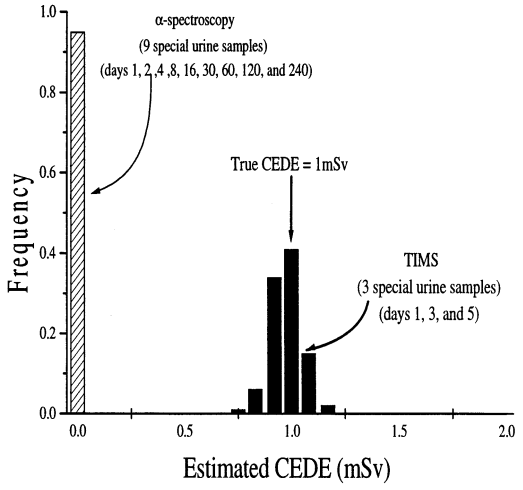

Fig. 4 Distributions from two 100 trial Monte Carlo experiments. Both distributions are based on an inhalation of 12 Bq, inhalation class *Y*, 1 μm AMAD, ^{239}Pu. This intake corresponds to a CEDE of 1 mSv. Results are based on an inhalation intake of class *Y*, 1 μm AMAD, ^{239}Pu. The biological variability used for both data sets was 10%.

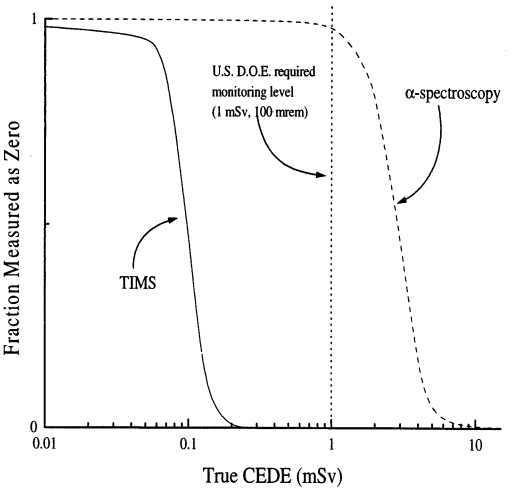

Fig. 5 Monte Carlo estimated fraction of intakes determined as zero intake for the indicated true CEDE for α spectroscopy and TIMS. Results are based on the collection of three 24 h urine samples at days 1, 3, and 5 after an inhalation intake of class *Y*, 1 μm AMAD, ^{239}Pu. The biological variability used for both data sets was 10%.

played in Fig. 4. The intake scenario would be expected to deliver a CEDE on the order of 1 mSv. Note the poor performance of the α-spectroscopy technique at this level. The results indicate a nearly 100% failure rate for detecting an acute intake resulting in 1 mSv CEDE, even if immediately followed up with an intensive nine sample urine assay program. The TIMS analysis reveals a distribution of CEDE estimates, with an expectation of 1 mSv and a population standard deviation of 0.1 mSv.

For the second case, Monte Carlo calculations were also performed over a range of committed effective dose equivalents (0.01 to 1000 mSv) for both the TIMS and α-spectroscopy analyses. In this case, results are based on the collection of three 24 h urine samples at days 1, 3, and 5 (for both measurement techniques) immediately after a known inhalation of class *Y*, 1 μm AMAD, ^{239}Pu. The biological variability used for both data sets was 10% and measurement bias was not included.

Results presented in Fig. 5 indicate the power of the TIMS measurement technique over α spectroscopy for monitoring plutonium workers. The TIMS results provide accurate estimates of CEDE down to the 0.2 mSv, compared to 5 mSv for α spectroscopy. The relative measurement errors provided by TIMS are also quite low when compared to α spectroscopy, as seen in Fig. 6. TIMS analysis provides a coefficient of variation on the order of 10% at 1 mSv CEDE.

An interesting feature of the TIMS technique, is the capability to obtain information regarding the presence and concentration of both ^{239}Pu and ^{240}Pu [18,20]. The ratio of ^{240}Pu atom to ^{239}Pu atom content in a sample reveals some information about the source of the plutonium [20]. For example, material used in the fabrication of the World War II nuclear weapons had a ratio of 0.01, weapons material in current use has a ratio of 0.06, and environmental material from open air testing has a ratio of 0.18. This information is applicable in establishing the approximate era and source of intake. Preliminary results indicate that this technique will identify statistically significant ratios when ^{239}Pu urine concentrations exceed 400 μBq per 24 h urine sample (approximately 5×10^8 atoms).

The TIMS analysis was implemented as an integral part of the LANL routine and special bioassay programs in January 1997. Individuals who handle plu-

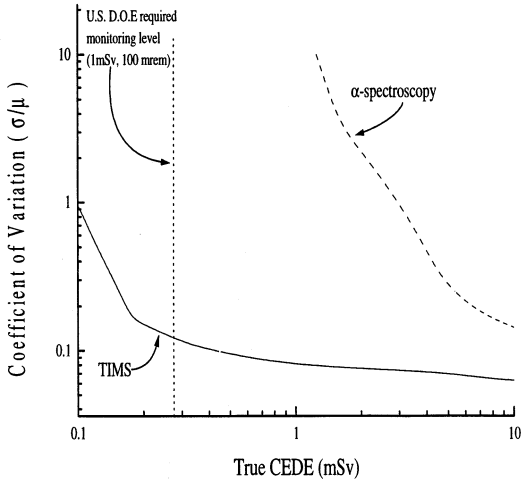

Fig. 6 Monte Carlo estimated relative error for average estimated intakes for the indicated true CEDE for α spectroscopy and TIMS. Results are based on the collection of three 24 h urine samples at days 1, 3, and 5 after an inhalation intake of class *Y*, 1 μm AMAD, ^{239}Pu. The biological variability used for both data sets was 10%.

tonium are routinely monitored through the collection of an annual 24 h urine sample. The sample is processed through the ultratrace chemistry procedure in class-100 clean rooms, and then is analyzed by both α spectroscopy and TIMS. Use of the α spectroscopy allows for a direct measure of chemical efficiency and the detection of ^{238}Pu in the urine sample. Implementation provides LANL with the ability to detect an intake resulting in a CEDE in the range of 1 to 3 mSv from a single annual routine urine sample, in the year of intake.

A special sampling program, termed *prompt* action, is required in cases where the CEDE is expected to exceed 10 mSv. This protocol utilizes three 24 h urine samples, collected over the five day period following an identified exposure situation. Additional samples may be collected at later times if necessary. For cases where the CEDE is expected to exceed 1 to 10 mSv, a single 24 h urine sample (*diagnostic action*) is collected following the exposure situation. Additional samples may be collected at later times if necessary. The current turnaround time to assess intakes from such exposure situations is three to six weeks. In emergency cases where the CEDE is expected to exceed 100 to 250 mSv, analysis may be performed in three to seven days. Detection of intakes with TIMS in the 10 to 20 Bq range and an associated CEDE of 0.5 mSv is possible under the special sampling program.

These analyses affirm the significance of lowering measurement uncertainties in order to more efficiently detect intakes of ^{239}Pu and ^{240}Pu. This capability has allowed LANL to approach the DOE regulation to monitor workers who may receive 1 mSv from the workplace. The combination of ultratrace chemistry performed in class-100 clean rooms and detection by thermal ionization mass spectrometry results in a superior bioassay result. Distributional analysis shows a relatively small bias. The intercomparison results verified that the LANL TIMS instrument is appropriate for application in the plutonium bioassay program for monitoring workers. The availability of a high quality estimate of the CEDE, in combination with a shorter turnaround time and fewer samples should provide the framework for better work place decision making, optimized monitoring cost benefit, and significant improvements in the ability to demonstrate compliance with DOE regulations.

Acknowledgements

We thank the following individuals for their assistance: Dr. R. J. Brake for the initial concept of application of mass spectrometry to occupational monitoring for plutonium in 1986 and for valuable technical discussions, D. E. Dry for alpha spectroscopy measurements, D. Lewis and Dr. M. E. Schillaci for valuable discussions in experimental planning and data analysis, Dr. H. Poths and J. Chamberlain for mass spectrometry measurements, F. R. Roensch, D. Kottmann, and C. Hensley for chemical separation of urine samples, C. Royer for data reduction and analysis, D. L. Wannigman for participant identification, sample collection, and valuable discussions on plutonium in the workplace, and the volunteers at the TA-55 Plutonium Facility for their time and effort in providing urine samples for this study. This project was funded by the Los Alamos National Laboratory, Environment, Safety, and Health Division, Technology Development and Engineering Applications program.

References

[1] Code of Federal Regulations 10, part 835, Office of the Federal Register, 1994.
[2] J.W. Healy, Ind. Hyg. Quart. 18 (1957) 261.
[3] J.N.P. Lawrence, Health Phys. 8 (1962) 61.
[4] W.D. Moss, E.E. Campbell, H.F. Schulte, G.L. Tietjen, Health Phys. 17 (1969) 571.
[5] S.R. Jones, Radiat. Prot. Dosim. 11, 1985.
[6] National Council on Radiation Protection and Measurements, Use of Bioassay for Assessment of Internal Radionuclide Deposition, National Council on Radiation Protection and Measurements, Bethesda, MD, 1987, NCRP Report No. 87.
[7] R.W. Leggett, K.F. Eckerman, Health Phys. 52 (1987) 337.
[8] International Commission on Radiological Protection, Recommendations of the International Commission on Radiological Protection, Individual Monitoring for Intakes of Radionuclides by Workers: Design and Interpretation, Vol. 19 of Annals of the ICRP, Pergamon, Oxford, 1988, ICRP Publication 54.
[9] B. Boecker, R. Hall, K. Inn, J.N.P. Lawrence, P. Ziemer, G. Eisele, B. Wachholz Jr., W. Burr, Health Phys. 60 (1991) 45, Suppl. 1.
[10] W.C.T. Inkret, G. Miller, Methods of Internal Dose Assessment, Technical Report ESH-12-02.01.0, Los Alamos National Laboratory, Los Alamos, NM, 1995.
[11] W.H. Langham, S.H. Bassett, P.S. Harris, R.E. Carter, Health Phys. 38 (1980) 1031.
[12] M. Gautier, Health and Environmental Chemistry: Analytical Techniques, Data Management, and Quality Assurance, Technical Report LA-10300-M, Los Alamos National Laboratory, Los Alamos, NM, 1983.
[13] G. Miller, W.C. Inkret, H.F. Martz, Radiat. Prot. Dosim. 58 (1995) 115.
[14] R. Toohey, E. Palmerand, L. Anderson, C. Berger, N. Cohen, G. Eisele, B. Wachholz Jr., W. Burr, Health Phys. 60 (1991) 7, Suppl. 1.
[15] D.E. Bihl, R.L. Buschbon, M.J. Sula, Health Phys. 65 (1993) 550.
[16] G. Miller, W.C. Inkret, M.E. Schillaci, Analyzing Bioassay Data Using Bayesian Methods, Technical Report, Los Alamos National Laboratory, Los Alamos, NM, 1997.
[17] Technology Shortfalls and Dose Determinations for Radioactive Material Intakes, 1995.
[18] R.E. Perrin, G.W. Knobeloch, V.M. Armijo, D.W. Efurd, Int. J. Mass Spectrom. Ion Processes 64 (1985) 17.
[19] D.W. Efurd, D.J. Rokop, R.D. Aguilar, F.R. Roensch, J.C. Banar, R.E. Perrin, Int. J. Mass Spectrom. Ion Processes 146 (1995) 109.
[20] D.W. Efurd, T.M. Benjamin, R.D. Aguilar, F.R. Roensch, R.E. Perrin, E.R. Gonzalez, J.F. McInroy, Measurement of Plutonium Activity Levels and $^{240}Pu/^{239}Pu$ Isotope Ratios in Bone Collected from Nonoccupationally-Exposed Individuals Living in Colorado, Technical Report LA-12952-MS, Los Alamos National Laboratory, Los Alamos, NM 87545, 1995.
[21] D.W. Efurd, D.J. Rokop, F.R. Roensch, Measurement of $^{240}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$ Atom Ratios in Soil Representative of Global Fallout in Colorado, Technical Report LA-UR-94-4200, Los Alamos National Laboratory, Los Alamos, NM 87545, 1994.
[22] M.E. Wrenn, M. Singh, M. Salamon, X.H. Xue, Plutonium Baseline Study in Human Urine, Technical Report, University of Utah, Salt Lake City, 1993, Progress Report to DOE.
[23] J.M.R. Hutchinson, Report of Traceability—^{239}Pu. Technical Report, National Institute of Standards and Technology, Washington, D.C., 1997, U.S. Department of Commerce.
[24] K.G.W. Inn, Intercomparison Study of Inductively Coupled Plasma Mass Spectrometry, Thermal Ionization Mass Spectrometry, and Fission Track Analysis of μBq Quantities of ^{239}Pu in syntheticUrine. Technical Report, National Institutes of Standards and Technology, Gaithersburg, MD 20899, August 1997, Report for Department of Energy Office of International Health Programs (EH-63).
[25] P.G. Johnson, R.D. Carver, R.J. Dupzyk, Health Phys. 33 (1977) 301.
[26] J.R. Moody, Anal. Chem. 54 (1982) 1358.
[27] National Council on Radiation Protection and Measurements, A Guide for Uncertainty Analysis in Dose Related to Environmental Contamination, National Council on Radiation Protection and Measurements, Bethesda, MD, 1996, NCRP Commentary No. 14.
[28] G. Miller, W.C. Inkret, H.F. Martz, Radiat. Prot. Dosim. 48 (1993) 251.
[29] International Commission on Radiological Protection, Limits for Intakes of Radionuclides by Workers, Annals of the ICRP, Pergamon, New York, 1979, ICRP Publication 30, Part 1.
[30] G. Miller, W.C. Inkret, Radiat. Prot. Dosim. 63 (1996) 189.
[31] G. Miller, W.C. Inkret, Bayesian Methods For Interpreting Plutonium Urinalysis Data, In Maximum Entropy and Bayesian Methods, Santa Fe, New Mexico, 1995, Kluwer Academic, Boston, 1996, pp. 367–373.