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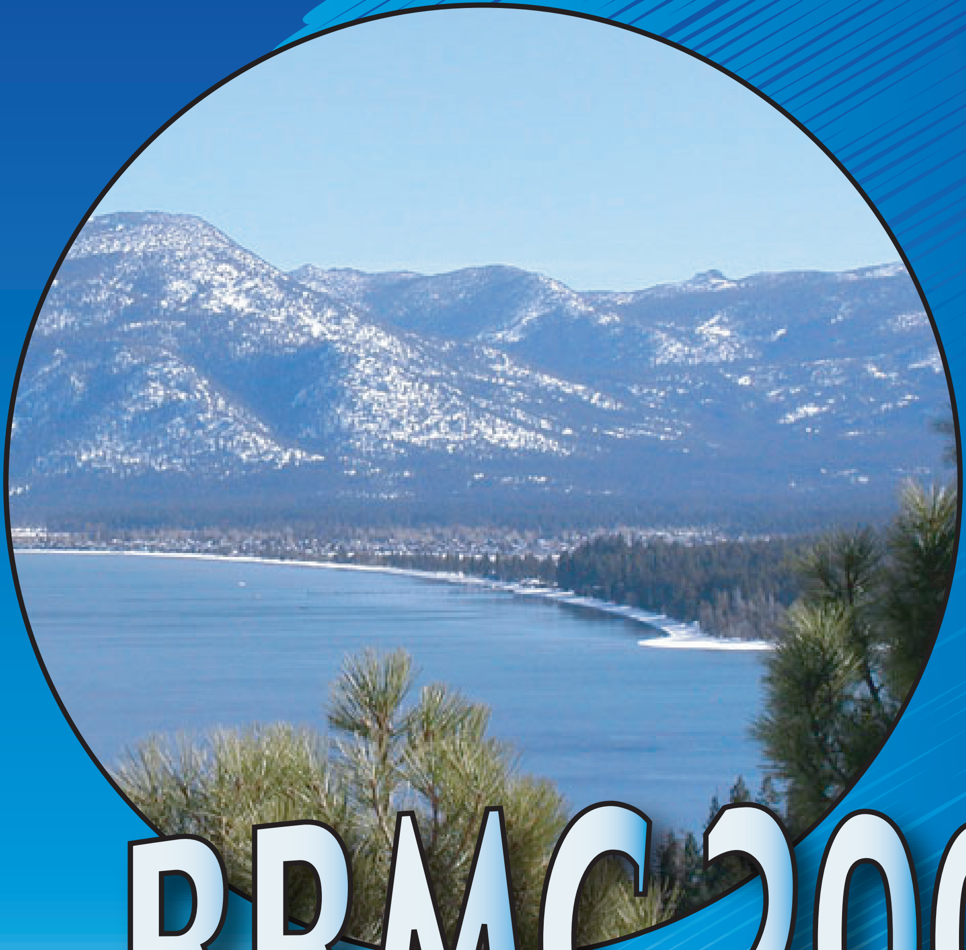

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# RRMC 2005

# 51st Annual
# Radiobioassay & Radiochemical Measurements Conference

C. Wong and G. Lane

October 24-28, 2005

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

**Welcome**

The Organizing Committee welcomes you to the 51st Annual Radiobioassay and Radiochemical Measurements Conference. At some time in our life each of us made a choice to pursue a career in radiochemistry. As we started on this path we began with questions that needed to be answered. In our efforts to answer these questions we built a foundation on which to base additional investigations. These investigations serve to advance the fields of radiobioassay and radiochemistry. At other times our ideas are challenged and what we once thought to be true must be rethought. By continuing to ask questions, pursuing new ways of doing things, and challenging ourselves, one soon finds that he or she has become an active part of this community.

Participants at the 51st Annual Radiobioassay and Radiochemical Measurements Conference represent all stages of careers in radiochemistry. As a new member to the radiochemistry community, some of the topics may be overwhelming. However, please ask your questions. Someone at this conference will likely have had a similar experience and would be willing to share their thoughts on the subject. For those of you further along in your careers, please participate by speaking up at a workshop, presenting a paper, joining a sub-committee or challenging the community to rethink what we thought to be true. By all means lend your expertise to augmenting the knowledge presented in papers and discussions with the intent of offering contributions, criticism and ideas for future research. This is your conference. Meet someone new, share your ideas, ask questions, and be challenged.

# 51st Annual
# Radiobioassay & Radiochemical Measurements Conference
# Schedule

# Workshop Schedule

|  | 10/24/2005 |  |  | 10/25/2005 |  |  |  |
|---|---|---|---|---|---|---|---|
|  | **Monday** |  |  | **Tuesday** |  |  |  |
| Breakfast 7:00 - 8:00 | Emperor Ballroom |  |  | Emperor Ballroom |  |  |  |
| Location | **Forum I** | **Forum II** | **Emperor Ballroom** | **Forum I** | **Forum II** | **Augustus** | **Emperor Ballroom** |
| 8:00 - 9:50 | **MARLAP Uncertainty** | **Ortec** |  | **CertusSoft** | **Canberra** | **DOELAP In Vitro** | **Vendor Displays**<br>**Poster Displays** |
| AM Break 9:50 - 10:10 | Emperor Ballroom |  |  | Emperor Ballroom |  |  |  |
| 10:10 - 12:00 | **MARLAP Uncertainty** | **Ortec** |  | **CertusSoft** | **Canberra** | **QA/QC** |  |
| Lunch 12:00 - 1:00 | On your own |  |  | On your own |  |  |  |
| 1:00 - 2:50 | **ICP/MS** | **Ortec** | Vendor Displays (Setup) | **Zumatrix** | **Eichrom** | **DOE Lung Intercomparison** |  |
| PM Break 2:50 - 3:10 | Forum I | Forum II |  | Emperor Ballroom |  |  |  |
| 3:10 - 5:00 | **ICP/MS** | **Ortec** |  | **NRIP** | **Protean** | **DOE Lung Intercomparison** |  |
| Evening |  |  |  | **Welcome Reception**<br>(Club Nero)<br>6:00 - 8:00 |  |  |  |

# Technical Session Schedule

| | 10/26/2005 | | | 10/27/2005 | | | 10/28/2005 |
|---|---|---|---|---|---|---|---|
| | **Wednesday** | | | **Thursday** | | | **Friday** |
| Breakfast 7:00 - 8:00 | Emperor Ballroom | | | Emperor Ballroom | | | Emperor Ballroom |
| Location | **Forum I / II** | **Augustus** | **Emperor Ballroom** | **Forum I / II** | **Augustus** | **Emperor Ballroom** | **Forum I / II** |
| 8:00 - 9:40 | **Welcome Keynote Speaker** | **Radiation - What is Important?**<br>**An Exhibit of Exempt Radioactive Items** | **Vendor Displays**<br>**Poster Displays** | **Bioassay *In Vitro*** | **Radiation - What is Important?**<br>**An Exhibit of Exempt Radioactive Items** | **Vendor Displays**<br>**Poster Displays** | **Radiometric Procedures** |
| AM Break 9:40 - 10:00 | Emperor Ballroom | | | Emperor Ballroom | | | Emperor Ballroom |
| 10:00 - 12:05 | **Standards QA/QC** | | | **Bioassay *In Vivo*** | | | **Radiometric Procedures** |
| Lunch 12:05 - 1:20 | On your own | | | On your own | Mark Hart Display (Tear down) | Vendor Displays (Tear down)<br>Poster Session (Tear Down) | **Lunch Keynote Speaker Business Meeting** (Circus Maximus) (rounds of 10) |
| 1:20 - 3:00 | **Mass Spectrometry** | **Radiation - What is Important?**<br>**An Exhibit of Exempt Radioactive Items** | | **Environmental Measurements** | | | |
| PM Break 3:00 - 3:20 | Emperor Ballroom | | | Forum I / II | | | Circus Maximus |
| 3:20 - 5:00 | **Poster Session** | | **ANSI 13.43** (TBD) **ASTM D19.04** (Senate) | **Standard Methods** (TBD) **ANSI N13.30** (Senate) | | | **Homeland Security** (Circus Maximus) |
| Evening | | | | Sunset Dinner Cruise<br>Tahoe Queen<br>5:00 - 9:30 | | | |

# Workshop Schedule

**Monday, October 24, 2005 - Forum I**

| | | | |
|---|---|---|---|
| 8:00 - 12:00 | Keith McCroan | MARLAP | MARLAP Uncertainty |
| 1:00 - 5:00 | Brad Esser and Ross Williams | ICP/MS | |

**Monday, October 24, 2005 - Forum II**

| | | | |
|---|---|---|---|
| 8:00 - 5:00 | Dave Martinez | Ortec | Alpha Spectrometry and Gamma Spectrometry |

**Tuesday, October 25, 2005 - Forum I**

| | | | |
|---|---|---|---|
| 8:00 - 12:00 | Heinrich Ziegler<br>Oleg B. Egorov | CertusSoft | Automation of Radioanalytical Chemistry |
| 1:00 - 2:50 | Rory Lewis | Zumatrix | Matrix Plus LMS for Radiochemistry Labs |
| 3:10 - 5:00 | Kenneth Inn | NRIP | NIST Radiochemistry Intercomparison Program Annual Users Meeting |

**Tuesday, October 25, 2005 - Forum II**

| | | | |
|---|---|---|---|
| 8:00 - 9:50 | Luc De Baerdemaeker Ing. BSc Msc | Canberra | QA/QC Implementation in Real World Gamma Spectroscopy Systems |
| 10:10 - 12:00 | Bill Cross | Canberra | Utilization of Ion Implanted Silicon Charged Particle Detectors in low Background Alpha / Beta Counting Systems for Radon, Thoron and progeny compensation |
| 1:00 - 2:50 | Larry Jassin | Eichrom | Radiochemical Methods |
| 3:10 - 5:00 | John Sonewald | Protean | Calibrating Alpha/Beta Counters with Cs-137 |

**Tuesday, October 25, 2005 - Augustus**

| | | | |
|---|---|---|---|
| 8:00 - 9:50 | Anita Bhatt | DOELAP | in Vitro |
| 10:10 - 12:00 | Mary Wisdom | QA | A Forum for Radiochemistry: QC and QA in the Real World |
| 1:00 - 5:00 | Tim Lynch | DOE | DOE Lung Intercomparison |

# Technical Program Schedule - Oral Presentations

**Wednesday, October 26, 2005 - Keynote Speaker**
**Chairperson: Carolyn Wong**

| | | | |
|---|---|---|---|
| 8:20 AM | Darleane Hoffman | University of California, Berkeley | The Crisis in Radiochemistry and Nuclear Chemistry Education |
| 9:00 AM | Mark Hart | University of California, Lawrence Livermore National Laboratory | Radiation - What is Important? An Exhibit of Exempt Radioactive Items |
| 9:25 AM | Virgene IdekerMulligan and Bob Shannon | Kaiser Analytical Management Services, Inc. | They Said It Couldn't Be Done… |

**Wednesday, October 26, 2005 - Standards & Standards Testing / QA / QC**
**Chairpersons: Bob Shannon & Matt Lardy**

| | | | |
|---|---|---|---|
| 10:10 AM | Donna M. Beals, David E. McCurdy, and Daniel E. Montgomery | Savannah River National Lab | Opportunities for the Advancement of Science Through Participation in the ASTM International |
| 10:35 AM | James Dahlgran, Mary Verwolf, David Sill, James Filliben, Gavin Spalletta, Kenneth G.W. Inn | RESL; NIST | Draft PT Uncertainty Acceptance Criteria Derived from Historical MAPEP Results |
| 11:00 AM | Cheryl Antonio | Pacific Northwest National Laboratory | Lessons Learned: Incidents Can Bring Unwelcome Surprises |
| 11:25 AM | Natasha Dimova, Bill Burnett and Henrieta Dulaiova | Environmental Radioactivity Measurement Facility, Department of Oceanography, Florida State University | Preparations of Radium Standards for Calibration of a Delayed Coincidence Counter System |
| 11:40 AM | T. L. Rucker | Science Applications International Corporation | Application of Decision Levels (DLs) and Minimum Detectable Amounts (MDAs) – Will the Madness Ever End? |

# Technical Program Schedule - Oral Presentations

**Wednesday, October 26, 2005 - Mass Spectrometry**
**Chairpersons: Dave McCurdy & Cindy Conrado**

| | | | |
|---|---|---|---|
| 1:20 PM | S. S. Kannamkumarath, G. R. Rao, B. D. Gonzalez, R. D. Myers, D. K. Hatmaker, and M. Thein | Oak Ridge Institute for Science and Education; Operational Safety Services Division Oak Ridge National Laboratory | An Improved Method for the Determination of $^{235}$U and $^{238}$U in Urine-By Inductively Coupled Plasma-Mass Spectrometry |
| 1:45 PM | Julian Wills, Meike Hamester, Chuck Douthitt | Thermo Electron | Application of extended dynamic range Sector Field ICP-MS to the analysis of high uranium matrices |
| 2:10 PM | Dominic Larivière, Stephen Kiser, Chunsheng Li, Vladimir Epov, R. Jack Cornett | Radiation Protection Bureau, Health Canada; Trent University | New and fast analytical protocol for the determination of actinides in air filter samples by HPLC-ICP-MS |
| 2:35 PM | Bogen, K.T., T.F. Hamilton, T.A. Brown, A.A. Marchetti, and R.E. Marinelli | University of California, Lawrence Livermore National Laboratory | Age-related trend in elevated plutonium-239 measured by AMS in urine samples collected in 1998-2003 from Enewetak residents and Rongelap resettlement workers |

# Technical Program Schedule - Oral Presentations

**Thursday, October 27, 2005 - Bioassay: In Vitro**
**Chairpersons: Gary Kramer & Sandi Fisher**

| | | | |
|---|---|---|---|
| 8:00 AM | Evan Crawford and Jerry LaRosa | Yale University; NIST | What a Coincidence-Developing a Rapid LSC Screening Method for $^{90}$Sr Contamination in Urine |
| 8:25 AM | John S. Morton, Robert P. Wills, Robert D. Timm, James B. Westmoreland | General Engineering Labs | On-site Collection, Concentration and Separation of Nuclide Specific Analytes in Urine |
| 8:50 AM | D. J. Fauth and T. R. La Bone | Westinghouse Savannah River Company | Validation Process for Bioassay MDCs |
| 9:15 AM | Bogen, K.T., D.P. Hickman, T.F. Hamilton, T.A. Brown, C.C. Cox, A.A. Marchetti and R.E. Martinelli | University of California, Lawrence Livermore National Laboratory | AMS Analysis of $^{239}$Pu in Archived Occupational Samples |

**Thursday, October 27, 2005 - Bioassay: In Vivo**
**Chairpersons: Stan Morton & Ilham AlMahamid**

| | | | |
|---|---|---|---|
| 10:00 AM | Gary H. Kramer and Barry M. Hauck | Human Monitoring Laboratory, Radiation Protection Bureau, Health Canada | The Human Monitoring Laboratory's new whole body counter, its comparison with the previous system and an extension of the calibration using Monte Carlo simulations |
| 10:25 AM | Matthew Mille and Svetlana Nour | University of Maryland; NIST | Getting into the BOMAB Head |
| 10:50 AM | Rachel A. Zeman, Sandra K. Fisher, David P. Hickman | University of California, Lawrence Livermore National Laboratory | The Heart Must be Removed Prior to In Vivo Lung Counting to Achieve Optimum Detection Efficiency of Low Energy Gammas |
| 11:15 AM | Malcolm Ennis, Andy Baker and Doug Van Cleef | Los Alamos National Laboratory; Advanced Measurement Technology, Inc | High Resolution HPGe Lung Counting Without Liquid Nitrogen |
| 11:40 AM | Gary H. Kramer and Barry M. Hauck | Human Monitoring Laboratory, Radiation Protection Bureau, Health Canada | The sliced BOMAB phantom: a new variant for intercomparisons. |

# Technical Program Schedule - Oral Presentations

**Thursday, October 27, 2005 - Environmental Measurements**
**Chairpersons: Bahman Parsa & Shiyamalie Ruberu**

| | | | |
|---|---|---|---|
| 1:20 PM | Mark A. Verdoorn, Dennis W. McBride | Idaho National Laboratory | Development of Autonomous Unmanned Vehicles for Radiation Surveys and Sampling |
| 1:45 PM | Abdul Khalique, Richard Pietz, and Richard Lanyon | Metropolitan Water Reclamation District of Greater Chicago | Radon Monitoring at the Metropolitan Water Reclamation District of Greater Chicago |
| 2:10 PM | Sherrod L. Maxwell | Westinghouse Savannah River Co. | Rapid Emergency Method for Actinides and Sr-89/90 in Water Samples |
| 2:35 PM | Tosheva Z.P., Kies A.F., Hofmann | University of Luxembourg | Investigation of radioactivity in waters in Luxemburg by follow-up radiochemical method |

# Technical Program Schedule - Oral Presentations

**Friday, October 28, 2005 - Radiometric Methods & Procedures**
**Chairpersons: Barry Stewart & Donna Beals**

| | | | |
|---|---|---|---|
| 8:00 AM | Cecilia C. DiPrete, Timothy A. DeVol, Christopher D. Theisen, and David P. DiPrete | Savannah River National Laboratory; Clemson University | Effect of Quench on Alpha/Beta Pulse Shape Discrimination of Liquid Scintillation Cocktails |
| 8:25 AM | Hiromu Kurosaki, Payam Motabar, and Ken Inn | National Institute of Standards and Technology; University of Maryland | Magic or Science? ~$^{90}$Sr efficiency calibration data reduction |
| 8:50 AM | J. La Rosa, I. Outola, E. Crawford | Ionizing Radiation Division, National Institute of Standards and Technology | Verification of $^{237}$Np activity concentration in a mixed radionuclides solution: recent experiences in chemical separation and measurement by alpha spectrometry |
| 9:15 AM | Shiyamalie R. Ruberu, Yun-Gang Liu, and S. Kusum Perera | California Department of Health Services | Occurrence and Distribution of $^{210}$Lead and $^{210}$Polonium in California Groundwater Determined by a Single Sample Analysis Method |

**Friday, October 28, 2005 - Radiometric Methods & Procedures**
**Chairpersons: Ken Inn & Ann Mullins**

| | | | |
|---|---|---|---|
| 10:00 AM | Payam Motabar, Svetlana Nour and Kenneth G. W. Inn | National Institute of Standards and Technology | Effect of Energy on Alpha-particle Counting Efficiency |
| 10:25 AM | M. Sadeghi, P. Van den Winkel, H. Afarideh and M. Haji-Saeid, L. De Vis, R. Waegeneer,A. De Schriiver. | Islamic Azad University; Center for Agriculture & Medicine (NRCAM); VUB-Cyclotron | Centrifugal electrodissolution system for irradiated rhodium target material used for the cyclotron $^{103}$Pd production |
| 10:50 AM | Barry Stewart | Carlsbad Environmental Monitoring and Research Center | Use of Thin Films to Control Recoil Contamination of Alpha Spectrometry Detectors |
| 11:15 AM | Edward Hall and Kenneth G.W. Inn | University of Rochester: NIST | Massive Impact, The Use of Collodion Films to Shield Solid State Alpha Detectors from Heavy Ion Recoils |
| 11:40 AM | Steven Wheland and Matt Lardy | STL Richland | Evaluation of Quench Corrected Liquid Scintillation Counting Instrument Blanks to Reduce False Positive Rate |

# Technical Program Schedule - Oral Presentations

**Friday, October 28, 2005 - Keynote Speaker**
**Chairperson: Bryan Bandong**

| | | | |
|---|---|---|---|
| 1:00 PM | Ken Moody | University of California, Lawrence Livermore National Laboratory | Discovery of Superheavy Elements |

**Friday, October 28, 2005 - Homeland Security / Emergency Response**
**Chairpersons: Bryan Bandong & Rod Melgard**

| | | | |
|---|---|---|---|
| 2:55 PM | Joel Swanson | University of California, Lawrence Livermore National Laboratory | The Radiological Assistance Program |
| 3:20 PM | Donna Beals and Doug Van Cleef | Savannah River National Lab; ORTEC | Using Existing Field Radiological Instrumentation to Influence Emergency Response Decisions |
| 3:45 PM | Gary H. Kramer | Human Monitoring Laboratory, Radiation Protection Bureau, Health Canada | Anatomy of an emergency exercise: EXFO. |
| 4:10 PM | Iisa Outola, Svetlana Nour, Hiromu Kurosaki, Jerry LaRosa, Kenneth G.W. Inn and Lynn Albin | NIST, MS 8462; Washington State Department of Public Health | Improvement in Radioassay Emergency Response Capability: NRIP'05 |
| 4:35 PM | Carolyn Wong, Robb Hadley, Mark Johnson, Dann Haynes | University of California, Lawrence Livermore National Laboratory | The Livermore HOTSPOT Mobile Laboratory |

# Technical Program Schedule - Poster Presentations

Display: Tuesday, October 25, 2005 - Thursday, October 27, 2005
Poster Session: Wednesday, October 26, 2005, 3:20 PM - 5:00 PM

| Poster Number | Authors/Co-Authors | Affiliation | Title |
|---|---|---|---|
| 1 | Sandra K. Fisher and David P. Hickman | University of California, Lawrence Livermore National Laboratory | Nuclear Accident Dosimeters at the Lawrence Livermore National Laboratory |
| 2 | Benjamin J. Hicks | Severn Trent Laboratories | Determination of Carbon-14 from Reactor Graphite |
| 3 | Benjamin J. Hicks, Joel Kempema, Steven L. Howard, Chelsea Jarrell | Severn Trent Laboratories; SAIC | Ra-226 by Alpha Spectroscopy, a Comparison of Preparation and Separation Methods by Two Independent Laboratories |
| 4 | Johnson, B.E., Esser B.K., Hunt, J.R., Conrado, C.L. and Guthrie, E | University of California, Lawrence Livermore National Laboratory | High resolution radionuclide analysis of sediments for reconstructing local releases |
| 5 | Gottfried Kueppers | Operation Management Nuclear Infrastructure, Department Decontamination (B-ND), Germandy | Fast source preparation procedures for alpha-spectrometry of the actinides |
| 6 | Dominic Lariviere, Ana Paula Packer, Chunsheng Li, Jing Chen, Jack Cornett | Radiation Protection Bureau, Canada | Age dependence of natural uranium and thorium concentration in human bone |
| 7 | P. E. Cisneros, C. J. Duffy, K. M. Israel, D. L. Kottmann, C. A. Lance, D. M. Lopez, L. J. Miller, S. B. Ortiz, J. L. Roach, F. R. Roensch, J. A. Weeks, R. E. Steiner, and S. P. LaMont | Isotope and Nuclear Chemistry Group, Los Alamos National Laboratory | Simplified Sample Preparation Chemistry for the Analysis of Plutonium by Thermal Ionization Mass Spectrometry (TIMS) |

# Technical Program Schedule - Poster Presentations

Display: Tuesday, October 25, 2005 - Thursday, October 27, 2005
Poster Session: Wednesday, October 26, 2005, 3:20 PM - 5:00 PM

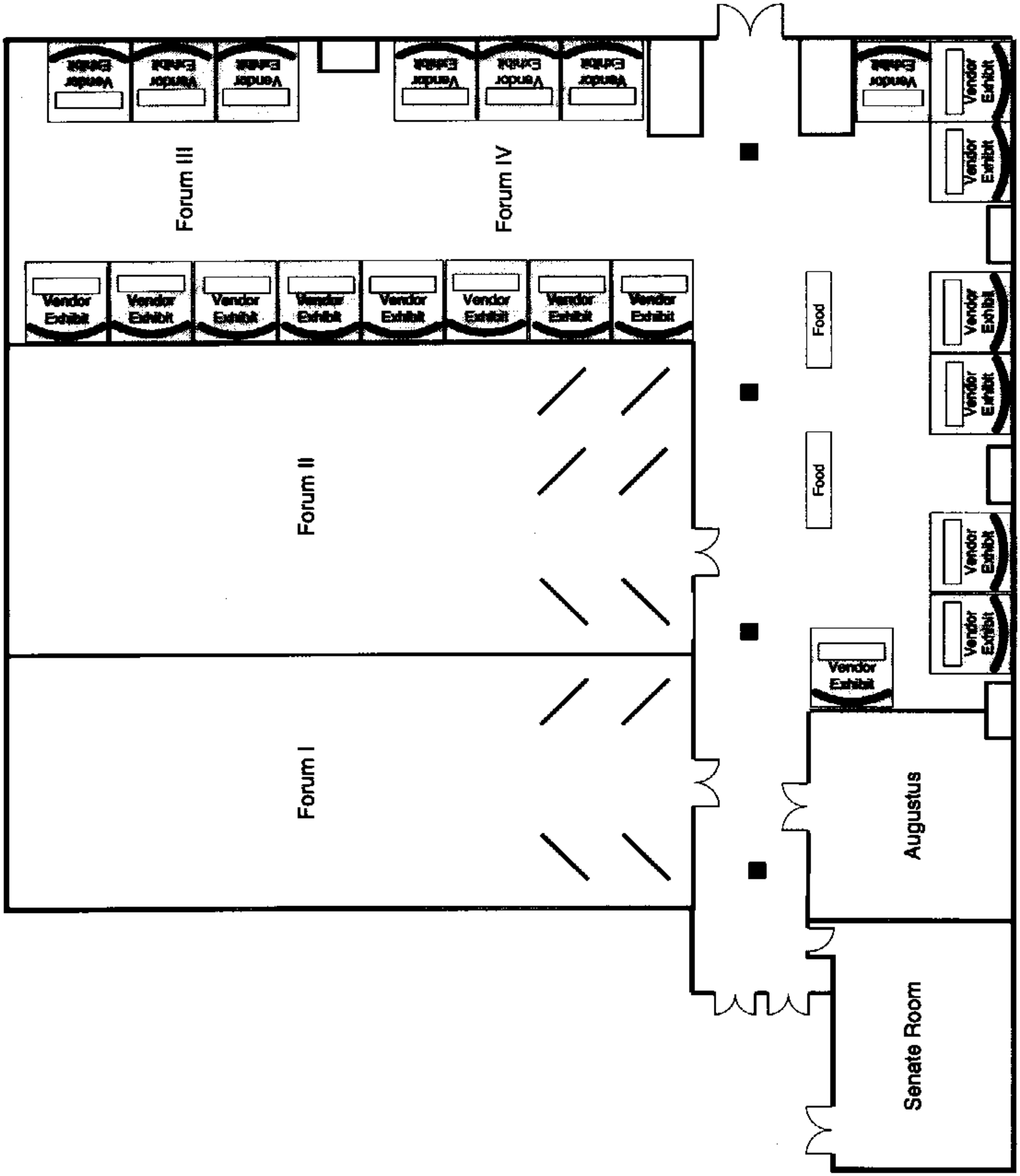
| Poster Number | Authors/Co-Authors | Affiliation | Title |
|---|---|---|---|
| 8 | Weijia Li, Chunsheng Li, Jiujiang Zhao, Stephen Kiser, Weihua Zhang, Dominic Lariviere, Jack Cornett | Radiation Protection Bureau, Health Canada; Carlton University | A Study toward Real Time Monitoring of Radionuclides in Fresh Waters Using Diffusive Gradients in Thin Films Technique Coupled with Germanium Gamma Ray Detector |
| 9 | Yun-Gang Liu, Shiyamalie R. Ruberu and S. Kusum Perera | California Department of Health Services | Gross Alpha /Beta Analyses of Groundwater by Liquid Scintillation Counting |
| 10 | H. K. Meznarich, E.d. Perez, K. Iwatate, D.G. Farwick | Flour Hanford | Technicium-99 Analysis at the Waste Sampling and Characterization Facility |
| 11 | Joseph Mwakapumba | Nevada State Health Laboratory | Sample pre-treatment and dissolution methods for analysis of radium-226, radium-228, and radium-224 by Liquid Scintillation Counter |
| 12 | A.M.G. Pacheco, M.C. Freitas, A. Machado, S. Sarmento, M.S. Baptista, M.T. Vasconcelos, J.P. Cabral | Technical University of Lisbon; Technological and Nuclear Institute; University of Port; Portugal | Vanadium accumulation in tree bark and epiphytic lichens at three sectors of Portugal |
| 13 | Arthur Scott and Philip Panter | Ontario Ministry of Labour - Radiation Protection Monitoring Service | Ontario Nuclear Reactor Surveillance Program – Tritium in Air |
| 14 | Donivan R. Porterfield, Lav Tandon, and Edward R. Gonzales | Los Alamos National Laboratory | Software Coincidence Spectrometry Using Commercially Available Instrumentation and Custom Developed Software |
| 15 | J. M. Rankin, G. F. Payne, and N.E. Bores | Oak Ridge National Laboratory | Intercomparison Studies Spikes Urine Samples used for Method Development and Validation |

# Technical Program Schedule - Poster Presentations

Display: Tuesday, October 25, 2005 - Thursday, October 27, 2005
Poster Session: Wednesday, October 26, 2005, 3:20 PM - 5:00 PM

| **Poster Number** | **Authors/Co-Authors** | **Affiliation** | **Title** |
|---|---|---|---|
| 16 | Bernd Kahn, <u>Robert Rosson</u>, Liz Thompson and Jeff Lahr | Georgia Tech Research Institute | Radioanalytical Chemistry |
| 17 | Donald E. Dry, <u>Joseph W. Sullivan</u> | Los Alamos National Laboratory | Advanced Analytical Data Handling for the Bioassay Program at LANL |

# 51st Annual
# Radiobioassay & Radiochemical Measurements Conference
# Exhibit Layout


Forum III
Forum IV
Forum II
Forum I
Augustus
Senate Room
Food
Food
Vendor Exhibit
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## Objectives

The objectives of the Conference (as adapted from the proceedings of the First Annual Bioassay and Analytical Chemistry Conference) are as follows:

- To bring everyone up-to-date on some of the latest developments in the field of bioassay, analytical and environmental radiochemistry;
- To enable all persons actively engaged in the field of bioassay, analytical and environmental radiochemistry to discuss mutual problems;
- To standardize some of the procedures commonly used by the various laboratories;
- To enable each laboratory to become familiar with procedures used elsewhere; and
- To plan for future meetings.

In recent years these objectives have been expanded to include the emerging field of Homeland Security.

These unique objectives provide a test crucible for pre-publication ideas, assumptions and approaches where the participants engage in intense straight-forward interactions to share new discoveries, challenge axioms, gain insights, share experiences, learn of the latest developments, seek solutions to technical problems and barriers, debate rationale, resolve technical issues, develop solutions and consensus, examine the impact of new policies and regulations, and establish valuable technical contacts. The Conference has been crucial to foster technical excellence and acts as a forum for the development of common but essential technical philosophies and practices that are used by federal and state government, national, contract, academic, and regulatory radioassay laboratories nationally and internationally.

In workshops, oral presentations and their defense, posters and discussions, technical workgroup sessions, and informal discussions, old and new colleagues meet to find out what are the latest, hottest developments amongst practitioners of bioassay, analytical and environmental radiochemistry. This is a unique Conference where the tricks-of-the-trade are revealed, new ideas are hatched, and the embryos of new collaborations are conceived.

## Conference Overview

This year's conference, as in the past, will consist of a series of vendor and technical workshops, oral presentations, poster presentations, vendor exhibits and related committee meetings. The conference is being organized to offer the participants full discussion of the oral presentations, ample opportunity to study the poster presentations, sufficient time to engage in in-depth discussions with each other and vendors, and flexibility to accommodate several workshops and related committee meetings.

Vendor and technical workshops will be held on Monday and Tuesday, and oral presentations will be given on Wednesday through Friday. Posters will be available for viewing on Tuesday through Thursday with a poster session on Wednesday afternoon. The vendor exhibits will be open from Tuesday through Thursday morning. We are very fortunate to have Mark and Jane Hart available with their display:

Radiation – What is Important
An Exhibit of Exempt Radioactive Items

This exhibit will be available for viewing on Wednesday and on Thursday morning.

The Conference this year is divided into eight topical areas, including the poster session:

| | |
|---|---|
| Standards & Standards Testing / QA / QC | Wednesday morning |
| Mass Spectrometry | Wednesday afternoon |
| Bioassay: In Vitro | Thursday morning |
| Bioassay: In Vivo | Thursday late morning |
| Environmental Measurements | Thursday afternoon |
| Radiometric Methods & Procedures | Friday morning |
| Homeland Security / Emergency Response | Friday afternoon |
| Poster Displays | Tuesday-Thursday |
| Poster Session | Wednesday late afternoon |

We have two keynote speakers scheduled to speak, the first on Wednesday during the opening session, and the second on Friday during the conference lunch. The business meeting will be held following lunch on Friday. The business meeting will confirm Chicago as the venue for the 2006 conference and accept offers for sponsoring the 2007 conference.

## History

The 51st Annual Radiobioassay and Radiochemical Measurements Conference is a continuation of an informal conference that has a long history. Over the past 51 years, the conference has also been known as:

- Annual Bioassay and Analytical Chemistry Conference,
- Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry,
- Annual Radiochemical Measurements Conference, and
- Annual Radiobioassay and Radiochemistry Measurements Conference

This Conference has become the most important forum where professionals can openly and informally discuss works-in-progress and common problems encountered during the development of state-of-the-art methods and procedures for detecting, measuring, and analyzing radioactive materials found in a large variety of environmental and biological media.

The Conference has had a rich history that has been influenced by leaders in the community who have been careful stewards of technical knowledge and philosophy and use the venue to encourage and guide openly and freely. The conference is attended by scientists from federal state and private laboratories, regulatory agencies, and equipment and service vendors. The hosting of the meeting is completely voluntary and has been sponsored by many different organizations over the years. The following is a history of the locations of the Conference:

| | |
|---|---|
| 1st – 1955 | Fernald, Ohio |
| 2nd – 1956 | Los Alamos, New Mexico |
| 3rd – 1957 | St. Louis, Missouri |
| 4th – 1958 | Rochester, New York |
| 5th – 1959 | Gatlinburg, Tennessee |
| 6th – 1960 | Santa Fe, New Mexico |
| 7th – 1961 | Argonne, Illinois |
| 8th – 1962 | Augusta, Georgia |
| 9th – 1963 | San Diego, California |
| 10th – 1964 | Cincinnati, Ohio |
| 11th – 1965 | Albuquerque, New Mexico |
| 12th – 1966 | Gatlinburg, Tennessee |
| 13th – 1967 | Berkeley, California |
| 14th – 1968 | New York, New York |
| 15th – 1969 | Los Alamos, New Mexico |
| 16th – 1970 | Bethesda, Maryland |
| 17th – 1971 | Boulder, Colorado |
| 18th – 1972 | Oakbrook Terrace, Illinois |
| 19th – 1973 | Moran, Wyoming |
| 20th – 1974 | Cincinnati, Ohio |
| 21st – 1975 | San Francisco, California |
| 22nd – 1976 | Philadelphia, Pennsylvania |
| 23rd – 1977 | Moran, Wyoming |
| 24th – 1978 | Annapolis, Maryland |
| 25th – 1979 | Las Vegas, Nevada |
| 26th – 1980 | Ottawa, Ontario, Canada |
| 27th – 1981 | Santa Fe, New Mexico |
| 28th – 1982 | Boston, Massachusetts |
| 29th – 1983 | Seattle, Washington |
| 30th – 1984 | Cincinnati, Ohio |
| 31st – 1985 | Chalk River, Ontario, Canada |
| 32nd – 1986 | Gaithersburg, Maryland |
| 33rd – 1987 | Berkeley, California |
| 34th – 1988 | Las Vegas, Nevada |
| 35th – 1989 | Charleston, South Carolina |
| 36th – 1990 | Oak Ridge, Tennessee |
| 37th – 1991 | Ottawa, Ontario, Canada |
| 38th – 1992 | Santa Fe, New Mexico |
| 39th – 1993 | Colorado Springs, Colorado |
| 40th – 1994 | Cincinnati, Ohio |
| 41st – 1995 | Boston, Massachusetts |
| 42nd – 1996 | San Francisco, California |
| 43rd – 1997 | Charleston, South Carolina |
| 44th – 1998 | Albuquerque, New Mexico |
| 45th – 1999 | Gaithersburg, Maryland |
| 46th – 2000 | Seattle, Washington |
| 47th – 2001 | Honolulu, Hawaii |
| 48th – 2002 | Knoxville, Tennessee |
| 49th – 2003 | Jackson Hole, Wyoming |
| 50th – 2004 | Cincinnati, Ohio |

# NOTES ON THE ANNUAL CONFERENCE ON BIOASSAY, ANALYTICAL, ENVIRONMENTAL RADIOCHEMISTRY

As Presented and Approved at the Business Meeting of the 38th Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry, Santa Fe, New Mexico, 1992

Suggestions to Promote Original Conference Objectives

## THE ENVIRONMENT

Every effort shall be made to maintain the environment for the annual meeting as an informal gathering of radiochemists, internal radiation dosimetrists, and those supporting such programs. To this end, the meeting host shall take steps to foster this goal that shall include:

1. Keep registration fees as low as possible;
2. Arrange hotel accommodations as near as possible to meeting areas and select accommodations that minimize costs to participants;
3. Schedule sessions to include some flexibility on length of papers as well as time for discussion (e.g. allowing time after each paper as well as scheduling general discussion periods after each session);
4. Adopt a meeting venue that fosters informal discussion between attendees;
5. Focus the meeting on technical presentations with vendor products and services as a secondary focus.

## THE MEETING

The annual meeting should be informal with the following guidelines:

1. Scheduled to start within the period from October 1 to October 31;
2. Scheduled in alternating years from eastern regions to western regions of North America, and for historical reasons, hosted every tenth year in the Cincinnati, Ohio area;
3. Technical papers to be scheduled over no more than a three (3) day period that shall be Tuesday through Thursday. Workshops, which may include panel discussions and vendor presentation, and local area tours of functions may be optionally arranged, at the host's discretion, on the Monday and/or Friday of that week;
4. Sessions to be moderated by appointed session chairperson so that discussion is directed to topics presented, but time allowed for papers is still loosely controlled so that each speaker is allowed the amount of time required to fairly address questions on their topic based on the potential impact of the information provided. The amount of time for each paper shall be variable based on the subject matter requirements;
5. Arrangements made for a business meeting which should, at least, cover the following:
    a. Soliciting volunteer hosts for the meeting to be held in two (2) years and confirming arrangements for the next year. If more than one candidate volunteers, the host will be selected from the vote of a simple majority of the attendees;
    b. Optionally, presenting meritorious service awards; and
    c. Opening up the forum to attendees for discussion.

## THE HOST

The hosting of the annual meeting shall be completely voluntary, but the volunteer host shall have several responsibilities that include:

1. Volunteering for hosting the meeting two (2) years in advance with prior approval of their corporate management. Vendors shall not be the sole host of conferences;
2. Arranging for the meeting rooms and/or visual supplies, and availability of hotel accommodations;
3. Maintaining, updating, and revision of the annual meeting attendee mailing list;
4. Sending out the following announcements:
    a. Meeting announcement with call for papers by March 1
    b. Final call for papers by July 1 (due August 1); and
    c. Preliminary meeting schedule with agenda by September 1
5. Arranging for advance advertising or announcement mailing to provide conference publicity at a level deemed appropriate by the host.
6. Arranging for printing the meeting proceedings that shall include extended abstracts (up to three (3) pages), final meeting schedule, list of historical meeting locations, and hosts, and list of past awardees of meritorious service awards;
7. Arranging the printing of an attendee list that should be disseminated during the conference;
8. Emphasizing the goal of the meeting as the meeting is opened. This shall include encouraging attendees to actively participate in discussion or openly asking questions;
9. Supporting and assisting with subcommittee and ad hoc committee meetings required by attendees;
10. Passing any surplus monies generated by the conference, finalized mailing lists, lists of meritorious service awardees, and this charter to the next host within three months of the last day of the meeting;
11. Optionally, selecting the winner of meritorious service award(s). Awardees shall have provided significant service to the technical field and the conference. These recipients and other attendees; and
12. Optionally, arranging for social "periods" for the benefit of conference attendees to be held between each day's sessions or after hours. The social "periods" or breaks may be sponsored by vendors or by other agencies on a voluntary basis.

Respectfully submitted by:

William Hayes, DataChem Laboratories
David McCurdy, Yankee Atomic Environmental Lab
Kenneth Inn, National Institute for Standard and Technology
Tracey Simpson, Lawrence Livermore National Laboratory
Gary Kramer, Bureau of Radiation and Medical Devices

# Founders Award

## HISTORY

The Organizing Committee of the 32nd Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry began the tradition by presenting three awards for meritorious service to the Bioassay Conference.

The Organizing Committees of several subsequent Annual Conferences carried on this tradition, which is not necessarily an annual event.

## THE AWARD

The award has taken the form of either a parchment, which was also signed by the attendees, or a wall plaque. The text of the award is as follows:

> **Whereas**, the participants in this conference pursue professional excellence and soulful cogitation,
>
> **Whereas**, topics of debate at this conference are at the forefront of scientific knowledge,
>
> **Whereas**, this conference is blessed with independent existence, and is dedicated to the preservation of free creativity and open exchange of intelligence, and
>
> **Whereas**, these publications of acclaim are the sole documents issued by this conference for public perlustration,
>
> **Be It Known**
>
> that all honors, rights, privileges, and praises which pertain to this letter of recognition are presented to
>
> **Name of Individual**
>
> by conference colleagues in sincere felicitation and appreciation for inspirational leadership and meritorious scientific contributions at the 51st Radiobioassay and Radiochemical Measurements Conference.

## THE RECIPIENTS

32nd Annual Meeting – Washington, DC
- Allen Brodsky
- Claude W. Sill
- Jake Sedlet

33rd Annual Meeting – Berkeley, CA
- Bill Moss
- Ken Heid
- Leon Leventhal

35th Annual Meeting – Charleston, SC
- David E. McCurdy
- Gary H. Kramer
- Kenneth G. W. Inn

37th Annual Meeting – Ottawa, Ontario
- Jim T. Harvey
- Roscoe M. Hall

38th Annual Meeting – Santa Fe, NM
- Jay E. McInroy

40th Annual Meeting – Cincinnati, OH
- Isabel M. Fisenne
- Robert Holtzmann

41st Annual Meeting – Boston, MA
- Myint Thein
- Henry B. Spitz
- Dale G. Olson

42nd Annual Meeting – San Francisco, CA
- Isabel Dupzyk
- Matt M. Lardy

43rd Annual Meeting – Charleston, SC
- Jack McDowell

44th Annual Meeting – Albuquerque, NM
- William C. Burnett
- David P. Hickman
- Daniel M. Montgomery
- John S. Morton

45th Annual Meeting – Gaithersburg, MD
- Bill Hayes
- Mo Khalil
- Jay MacLellan
- Tom Rucker

46th Annual Meeting – Seattle, WA
- Lee Sygitowicz
- Chuan Fu Wu
- Bahman Parsa
- Amir Mohaghaghi
- Robert Loesch

47th Annual Meeting – Honolulu, HI
- Donivan R. Porterfield

48th Annual Meeting – Knoxville, TN
- Mary Wisdom
- Michael Fern

49th Annual Meeting – Jackson Hole, WY
- Sandra Wagner
- Rodney Melgard
- Robert Shannon
- Donna Beals

50th Annual Meeting – Cincinnati, OH
- Norman Cohen
- Richard Blanchard
- Naomi Harley

## Acknowledgements

The Organizing Committee recognizes the efforts of numerous individuals and organizations that have made this conference possible. We acknowledge the support of the vendors who provide many of the extras that are greatly appreciated. We urge the participants to make time during this conference to visit the Vendor area and express your thanks for their support.

This conference would not have been possible without the support of the sponsors. The efforts of the conference organizing committee were supported by the LLNL Hazards Control Department, Chemical, Biology and Nuclear Sciences Division, Atmospheric, Earth, and Energy Department and Lawrence Berkeley National Laboratory. Eberline Services also provide invaluable support for the conference.

But, most of all, the committee appreciates and values the participants of the conference, no matter the extent.

## Organizing Committee

| | |
|---|---|
| Conference Co-Chairs: | Carolyn Wong (LLNL)<br>Cindy Conrado (LLNL) |
| Administrative Support: | Gina Lane (LLNL) |
| Technical Program Chair: | Bryan Bandong (LLNL) |
| Technical Workshops: | Carolyn Wong (LLNL) |
| Vendor Liaison: | Rodney Melgard (Eberline Services) |
| Organizing Committee Members: | Ilham AlMahamid (LBL)<br>Rayla Bradsher (LLNL)<br>Richard Bibby (LLNL)<br>Lori Collins (LLNL)<br>Brad Esser (LLNL)<br>Terry Hamilton (LLNL)<br>Dave Hickman (LLNL)<br>Hazel Holloway (LLNL)<br>Cherie Napier (LLNL) |
| Website Design: | Marisa Price (LLNL) |

# Vendor Listing

| Vendor | Contact |
|---|---|
| **Analytics Inc.**<br>1380 Seaboard Industrial Blvd.<br>Atlanta, GA 30318<br>USA | Daniel Montgomery<br>Voice: 404) 352-8677<br>Fax: (404) 352-2837<br>Email: customerservice@analyticsinc.com<br>Web: http://www.analyticsinc.com |
| **Canberra Industries**<br>800 Research Parkway<br>Meriden, CT 06450<br>USA | Tammy Pattison<br>Voice: (203) 639-2148<br>Fax: (203) 235-1347<br>Email: tpattison@canberra.com<br>Web: http://www.canberra.com |
| **CEM**<br>3100 Smith Farm Road<br>Matthews, NC 28106<br>USA | Michael Burnett<br>Voice: (800) 726-3331 ext 424<br>Fax: 925) 755-9865<br>Email: Michael.Burnett@cem.com<br>Web: http://www.cem.com |
| **CertusSoft Ltd.**<br>37 Kings Road<br>Brentwood, Essex<br>CM14 4DJ<br>UK | Michaela Lenz<br>Voice: ++! (519) 685-6665<br>Fax: ++1 (519) 685-6665<br>Email: Michaela.Lenz@CertusSoft.com<br>Web: http://www.certussoft.com |
| **Chemcheck**<br>1845 Terminal Dr<br>Suite 101<br>Richland, WA 99354-4959<br>USA | Kaelen Sanders<br>Voice: (509) 943-5000<br>Fax: (509) 946-3368<br>Email: chemchek@earthlink.net<br>Web: http://www.chemchek.com |
| **Eberline Services**<br>2030 Wright Ave.<br>P.O. Box 4040<br>Richmond, CA 94804-0040<br>USA | Rodney K. Melgard<br>Voice: (510) 235-2633<br>Fax: (510) 235-0438<br>Email: rmelgard@eberlineservices.com<br>Web: http://www.eberlineservices.com |
| **Eichrom Technologies, Inc.**<br>8205 South Cass Ave.<br>Suite 106<br>Darien, IL 60561<br>USA | Lawrence Jassin<br>Voice: (630) 963-0320<br>Fax: (630) 963-1928<br>Email: info@eichrom.com<br>Web: http://www.eichrom.com |

# Vendor Listing

| Vendor | Contact |
|---|---|
| **Environmental Express**<br>490 Wando Park Blvd.<br>Mt. Pleasant, SC 29464<br>USA | Paul Strickler<br>Voice: (843) 881-6560<br>Fax: (843) 881-3964<br>Email: pauls@envexp.com<br>Web: http://www.environmentalexpress.com |
| **Environmental Inc. Midwest Laboratory**<br>700 Landwehr Road<br>Northbrook, IL 60062<br>USA | Bronia Grob<br>Voice: (847) 564-0700<br>Fax: (847) 564-4517<br>Email: bgrob@midwestlaboratory.com<br>Web: http://www.alleghenytechnologies.com/wahchang/ |
| **Gamma Products, Inc.**<br>7730 W. 114th Place<br>Palos Hills,IL 60465<br>USA | Walter Meier<br>Voice: (708) 974-4100<br>Fax: (708) 974-0071<br>Email: sales@gammaproducts.com<br>Web: http://www.gammaproducts.com |
| **General Engineering Laboratories, LLC**<br>2040 Savage Road<br>Charleston, SC 29407<br>USA | Stan Morton<br>Voice: (303) 349-8345<br>Fax:<br>Email: stan.morton@gel.com<br>Web: http://www.gel.com |
| **Laboratory Data Consultants, Inc.**<br>7750 El Camino Real, Suite 2L<br>Carlsbad, CA 92009<br>USA | Richard Amano<br>Voice: (760) 634-0437<br>Fax: (760) 634-1674<br>Email: ramano@lab-data.com<br>Web: http://www.lab-data.com |
| **Ordela, Inc.**<br>1009 Alvin Weinberg Drive<br>Oak Ridge, TN 37830<br>USA | Wayne P. Graves<br>Voice: (865) 483-8675<br>Fax: (865) 483-8404<br>Email: WPGraves@ordela.com<br>Web: http://www.ordela.com/ |

# Vendor Listing

| **Vendor** | **Contact** |
|---|---|
| **ORTEC**<br>801 South Illinois Ave<br>Oak Ridge, TN 37831<br>USA | Susie Brockman<br>Voice: (865) 483-2124<br>Fax: (865) 425-1380<br>Email: info@ortec-online.com<br>Web: http://www.ortec-online.com |
| **Paragon Analytics**<br>225 Commerce Drive<br>Ft. Collins, CO 80524<br>USA | Claudia Clark<br>Voice: (207) 422-1506<br>Fax: (270) 422-1507<br>Email: cclark@bbtel.com<br>Web: http://www.paragonlabs.com |
| **PerkinElmer Life and Analytical Sciences**<br>2200 Warrenville Rd.<br>Downers Grove, IL 60515<br>USA | Chuck Passo<br>Voice: (630) 322-5400<br>Fax: (630) 322-5207<br>Email: chuck.passo@perkinelmer.com<br>Web: http://www.perkinelmer.com |
| **Princeton Gamma-Tech Instruments**<br>1026 Route 518<br>Rock Hill, NJ 08553<br>USA | Richard Varall<br>Voice: (609) 924-7310 ext: 306<br>Fax: (609) 924-1729<br>Email: rvarall@ptg.com<br>Web: http://www.pgt.com |
| **Protean Instrument Corporation**<br>231 Sam Rayburn Parkway<br>Lenoir City, TN 37771<br>USA | John Sonewald<br>Voice: (865) 717-3456<br>Fax: (865) 717-3456<br>Email: jsonewald@proteaninstrument.com<br>Web: http://www.proteaninstrument.com |
| **Severn Trent Laboratories, Inc. (STL)**<br>2800 George Washington Way<br>Richland, WA 99354<br>USA | Marty Cahill<br>Voice: (509) 375-3131<br>Fax: (509) 375-5590<br>Email: MCahill@stl-inc.com<br>Web: http://www.stl-inc.com |

# Vendor Listing

| Vendor | Contact |
|---|---|
| **Teledyne Brown Engineering**<br>2508 Quality Lane<br>Knoxville, TN 37931<br>USA | Rebecca L. Charles<br>Voice: (865) 934-0379<br>Fax: (865) 934-0396<br>Email: Rebecca.charles@tbe.com<br>Web: http://www.tbe.com/products/radiological_lab.asp |
| **Zumatrix, Inc.**<br>29 Simpson Lane<br>Unit D1<br>Falmouth, MA 02540<br>USA | Peter Mansfield<br>Voice: (508) 457-7911<br>Fax: (508) 457-7993<br>Email: info@zumatrix.com<br>Web: http://www.zumatrix.com |

# 51st Annual
# Radiobioassay & Radiochemical Measurements Conference
# Workshop Sessions

# Workshop Schedule

**Monday, October 24, 2005 - Forum I**

| | | | |
|---|---|---|---|
| 8:00 - 12:00 | Keith McCroan | MARLAP | MARLAP Uncertainty |
| 1:00 - 5:00 | Brad Esser and Ross Williams | ICP/MS | |

**Monday, October 24, 2005 - Forum II**

| | | | |
|---|---|---|---|
| 8:00 - 5:00 | Dave Martinez | Ortec | Alpha Spectrometry and Gamma Spectrometry |

**Tuesday, October 25, 2005 - Forum I**

| | | | |
|---|---|---|---|
| 8:00 - 12:00 | Heinrich Ziegler<br>Oleg B. Egorov | CertusSoft | Automation of Radioanalytical Chemistry |
| 1:00 - 2:50 | Rory Lewis | Zumatrix | Matrix Plus LMS for Radiochemistry Labs |
| 3:10 - 5:00 | Kenneth Inn | NRIP | NIST Radiochemistry Intercomparison Program Annual Users Meeting |

**Tuesday, October 25, 2005 - Forum II**

| | | | |
|---|---|---|---|
| 8:00 - 9:50 | Luc De Baerdemaeker Ing. BSc Msc | Canberra | QA/QC Implementation in Real World Gamma Spectroscopy Systems |
| 10:10 - 12:00 | Bill Cross | Canberra | Utilization of Ion Implanted Silicon Charged Particle Detectors in low Background Alpha / Beta Counting Systems for Radon, Thoron and progeny compensation |
| 1:00 - 2:50 | Larry Jassin | Eichrom | Radiochemical Methods |
| 3:10 - 5:00 | John Sonewald | Protean | Calibrating Alpha/Beta Counters with Cs-137 |

**Tuesday, October 25, 2005 - Augustus**

| | | | |
|---|---|---|---|
| 8:00 - 9:50 | Anita Bhatt | DOELAP | in Vitro |
| 10:10 - 12:00 | Mary Wisdom | QA | A Forum for Radiochemistry: QC and QA in the Real World |
| 1:00 - 5:00 | Tim Lynch | DOE | DOE Lung Intercomparison |

# MARLAP Measurement Uncertainty Workshop

Keith D. McCroan, U.S. Environmental Protection Agency
Kenneth G.W. Inn, National Institute of Standards and Technology

Presenters: William F. Guthrie
Robert Litman
Keith D. McCroan
David E. McCurdy
Thomas W. Vetter

The *Multi-Agency Radiological Analytical Protocols Manual* (MARLAP), which was approved and released in 2004, recommends a performance-based approach to planning and implementation of projects that involve radiochemical analysis. Measurement uncertainty plays a crucial role in much of the manual's guidance. In particular, MARLAP's approach to method selection in Part I is based on the planning team's uncertainty requirements for decision-making, and criteria for evaluating the continuing performance of the laboratory are based on the "required method uncertainty," a concept introduced by MARLAP. For these reasons, Part II of MARLAP, which is aimed at laboratory personnel, emphasizes the need for good uncertainty evaluations. MARLAP recommends that labs follow the *Guide to the Expression of Uncertainty in Measurement* (the "GUM") to ensure consistency and comparability of uncertainty statements.

This workshop provides an introduction to the role of measurement uncertainty in the MARLAP guidance and demonstrates methods and tools that can be used to make uncertainty evaluations in the laboratory easier. Specific topics of the workshop include:

- an overview of the role of uncertainty in MARLAP
- a brief refresher on the GUM
- the definition of the "required method uncertainty" and its role in selecting a method and evaluating a lab's continuing performance
- the use of software tools for propagating uncertainty automatically, including the Kragten spreadsheet (*Analyst*, **119**, 1994, 2161—2165), software component libraries, and standalone applications

# The Kragten Spreadsheet: An Easier Way to Estimate Uncertainty

Thomas W. Vetter, NIST Analytical Chemistry Division
William F. Guthrie, NIST Statistical Engineering Division

A spreadsheet tool based on the Kragten spreadsheet (*Analyst*, **119**, 1994, 2161-2165), that simplifies the calculation of the combined standard uncertainty, $u_c$, is described. No calculus is required. Given a measurement equation and mean values, standard uncertainties, and degrees of freedom for each input, this spreadsheet automatically calculates the combined standard uncertainty, effective degrees of freedom, coverage factor, and expanded uncertainty. In addition, the spreadsheet computes additional quantities, such as sensitivity coefficients and relative contributions of each uncertainty component to the combined standard uncertainty, which can give insight into the measurement process and can be used for optimization. The concept of the Kragten spreadsheet, as well as its advantages and disadvantages, compared to using calculus or relative uncertainties, are explained. The utility of the spreadsheet and steps to ensure the validity of its results are demonstrated with examples.

**ORTEC Technical Workshop for Monday, Oct. 24, 2005**

8:30 Welcome - Doug Van Cleef - Manager,ORTEC Distribution and Technical Support, Americas and Benson Davis- Systems Sales and Technical Support, Advanced Measurement Technology - ORTEC

8:45 "Basic Physics for Alpha and Gamma Spectroscopy", Doug Van Cleef, Manager ORTEC Distribution and Technical Support, Americas
This introduction includes a review of the nature and origins of gamma and alpha radiation as well as basic physics of interaction with matter.

9:45 "Fundamentals of Alpha Spectroscopy", Craig Maddigan, Application Specialist, Advances Measurement Technology, ORTEC
2005-00-044* Fundamentals of Alpha Spectroscopy (2 hours)
This course offers a fast-paced review of the basic principles of alpha spectroscopic analysis. The course includes a review of the considerations and consequences of sample preparation for alpha spectroscopy, alpha spectroscopy system components and calibrations, and a primer on interpretation of alpha spectroscopy data. This course is two hours in duration (when combined with the Basic Physics course above) and the American Academy of Health Physics will grant 4 continuing education credits for completion.

10:45 Break

11:00 "Fundamentals of Gamma Spectroscopy", Benson Davis - Technical Support and Systems Sales Support Advances Measurement Technology - ORTEC.
2005-00-043* Fundamentals of Gamma Spectroscopy (2 hours)
This course offers a fast-paced review of the basic principles of gamma spectroscopic analysis. The course includes a review of the nature and origins of gamma-emitting radioactivity, basic physics of gamma interaction with matter, consequences of gamma interactions on gamma spectra, gamma spectroscopy system components and calibrations, gamma spectroscopy analysis methods and interpretation of gamma spectroscopy data. The course is two hours in duration (when combined with the Basic Physics course above) and the American Academy of Health Physics will grant 4 Continuing Education Credits for completion.

12:00 Lunch

1:15 - 2:00 1st User Presentation (Gary Kramer - Health Canada). Human Monitoring Laboratory

2:00 - 2:45 2nd User Presentation (Henrieta Dulaiova - Environmental Radioactivity Measurement Facility, Florida State University) - The advantage of the use of low background gammaspectrometry for counting 226Ra and 228Ra in natural water samples.
Abstract: One of the methods that we use for analysis of radium in natural waters is by preconcentration via adsorption onto MnO2-coated acrylic fiber. The fiber containing radium is ashed and sealed into a crucible which is counted by gamma-spectrometry. 228Ra is determined by 228Ac (338 and 911 keV), 226Ra is determined either by its direct photopeak at 186 keV

(clear from interferences from 235U) or by counting 214Bi (609 keV) and 214Pb (295 and 351 keV). It is important to use a low background counting system since radionuclides from building structures and primordial emitters from the cryostat may contribute significantly to the backgrounds of the peaks of interest.

The gamma-spectrometry system in our facility at FSU consists of a GMX series detector ( 16.4 % at 1332 keV), housed in a 4" Pb shielding with 0.02"Cd and 0.06" Cu liners in the cavity. The full-energy peak backgrounds measured with blank fiber were 0.10 cpm at 295 keV, and 0.14 cpm at 609 keV. The calculated MDA for 226Ra for a 20-liter water sample and 2 days of counting is 0.7 dpm L-1 via the 186 keV peak, ~0.23 dpm L-1 (295, 352, 609 keV), and ~0.22 dpm L-1 for 228Ra.
A set of radium fibers was counted on Ortec's low background gamma-spectrometric system. The system consists of a GEM FX (47.3% at 1332 keV) series detector in a Pop-Top capsule configuration. The shielding has 2-6" Pb walls, 0.06" Sn and 0.25" Cu lining, with additional low background shielding in the capsule. The backgrounds of the blank fiber were 0.006 cpm at 295 keV, and 0.008 cpm at 609 keV. The lower background and higher efficiency of this detector provided a much lower MDA than the FSU system. This allowed us to reach the same MDA in only5-6 hours. The low background system was much more effective due to a better sample/detector geometry, higher efficiency, but mainly because of almost two orders of magnitude lower backgrounds. This improvement allowed us to count 50 samples in only 25 days rather than the ~75 days on our own system.

2:45 - 3:00 Break

3:00 - 3:45 3rd User Presentation (Don Dry - LANL) - LANL's Experience with AlphaVision
3:45 - 4:30 4th User Presentation (Jennifer Hartel - CDC) - "Incorporating a Radionuclide laboratory into an inorganic laboratory; trials and tribulations"
Abstract: For over 30 years our laboratory has focused on the periodic table for analytes of interest to pursue for public health applications. Recently under the directive of the Federal Response Plan - Emergency Support Function (ESF) #8, our role at the Centers for Disease Control and Prevention is to rapidly evaluate human exposure to a radiological event. Currently we our focusing our efforts on developing a rapid gamma spectrometry method for detection of various nuclides.
4:30 Questions / Discussions

# Fundamentals of Alpha Spectroscopy

Abstract: This course offers a fast-paced review of the basic principles of alpha spectroscopic analysis. The course includes a review of the nature and origins of alpha-particle emitting radioactivity, basic physics of alpha particle interaction with matter, considerations and consequences of sample preparation for alpha spectroscopy, alpha spectroscopy system components and calibrations, and a primer on interpretation of alpha spectroscopy data. The course is two hours in duration and the American Academy of Health Physics will grant 4 Continuing Education Credits for completion. There is no cost for attendance at this seminar.

Presented by: ORTEC/Advanced Measurement Technology, Inc., Oak Ridge, TN

# Fundamentals of Gamma Spectroscopy

Abstract: This course offers a fast-paced review of the basic principles of gamma spectroscopic analysis. The course includes a review of the nature and origins of gamma-emitting radioactivity, basic physics of gamma interaction with matter, consequences of gamma interactions on gamma spectra, gamma spectroscopy system components and calibrations, gamma spectroscopy analysis methods, and interpretation of gamma spectroscopy data. The course is two hours in duration and the American Academy of Health Physics will grant 4 Continuing Education Credits for completion. There is no cost for attendance at this seminar.

Presented by: ORTEC/Advanced Measurement Technology, Inc., Oak Ridge, TN

# CertusSoft Workshop on
# Automation of radioanalytical chemistry

Workshop organized by CertusSoft. Ltd, UK, www.certussoft.com
Chairperson: Dr. Heinrich Ziegler, Heinrich.Ziegler@CertusSoft.com
Co-chair: Dr. Oleg B. Egorov, PNNL, USA; oleg.egorov@pnl.gov

**Abstract:** Broad range radioanalytical measurements are carried out in support of commercial nuclear applications, nuclear waste processing, environmental monitoring, cleanup and restoration of the contaminated sites, verification of nonproliferation treaties, and other national and global security applications. Wet radiochemical analysis techniques continue to be paramount importance in the analysis of beta and alpha emitting radionuclides that can be readily measured using gamma or mass spectroscopic techniques. In many cases radioanalytical methods require tedious and labor intensive sample preparation, separation, detection and data interpretation procedures. Often times baseline radioanalytical procedures are based on outdated and inefficient methodologies. Furthermore, manual techniques continue to be dominant in radioanalytical laboratories even in the cases where the benefits of automation are obvious.

Comprehensive laboratory automation is becoming a main stream in clinical and bioanalytical laboratories. Recent advancements in separation materials, analytical instrumentation, automated fluid handling techniques, laboratory robotics, and computer hardware and software provide a solid foundation for the development of automated radioanalytical techniques. And yet, radiochemists continue to be confronted with the statements "This can not be automated…".

The purpose of this workshop is to provide an insight into the possibilities of automation for various tasks performed in radiochemical laboratories. Representatives of CertusSoft, dedicated to providing custom automation solutions, and scientists from national laboratories will discuss:

1) automation of separation techniques with robots;
2) automation of separation techniques with HPLC;
3) automated radionuclide analyzers and water monitoring sensors;
4) automation of data processing and evaluation;
5) extending third party solutions to improve efficiency and user friendliness;
6) capturing and making available knowledge and techniques of experienced scientists using computer-based knowledge bases (semi-intelligent databases);

CertusSoft Ltd. has been founded to deliver custom made automation solutions where others have failed or have not even tried. The company philosophy is "*Our customer's increased success in business is our success*". Dr. Ziegler, radiochemist and professional software developer / software architect, will share his automation experience. Part of this experience he gained working five years for the laboratories of the IAEA (Vienna) and ITU (Karlsruhe). Working four years in the pharmaceutical industry provided an insight into high throughput automation, professional database design and professionalism in software development / architecture in a highly demanding industry.

Dr, Egorov, a Senior Research Scientist at the Pacific Northwest National Laboratory will share results of the decade long research on automated radionuclide analysis and sensing at the US DOE Hanford site.

ZUMATRIX

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## Abstract for Vendor Workshop at RRMC 2005

## Matrix Plus LIMS for Radiochemistry Labs

Presenter: Rory Lewis, Zumatrix, Inc.

The processing, tracking, managing and reporting of samples, tests and results in a radiochemistry laboratory requires specialization beyond standard commercial LIMS software. There are requirements that are common between most radiochemistry labs and there are those that are specific to individual labs. To accommodate both these unique and common requirements Zumatrix has a configuration of Matrix Plus specifically for radiochemistry labs and the tools that allow system configuration to meet individual lab demands without the need to write custom code or scripts. It is this combination of a commercial LIMS with the base radiochemistry configuration and the powerful configuration tools that delivers the benefits of commercial software with the fit of custom in-house developments. This approach has been very successful with the industry in the UK and has a growing recognition in North America.

The presentation will highlight the benefits and capabilities of Matrix Plus for this market and the flexibility to meet the differing requirements between labs.

# NRIP Workshop

# NIST Radiochemistry Intercomparison Program

# Annual Users Meeting

In 1997 the Radioactivity Group established a traceability-testing program for low-level radiochemistry measurements. Each year, four rounds of evaluations are conducted with participating university, federal, national, interest group, and contract laboratories. The matrices (water, soil, sediment, air filter, synthetic feces and synthetic urine) and activity concentrations reflected common radioanalytical analyses performed by the participating laboratories. In addition, since 2004, NRIP has also included an Emergency Radiological Preparedness component to the program.

Laboratories are issued Certificates of Traceability for the evaluation results. The program has been implemented to meet the guidance for traceability as defined under ANSI N42.23, ANSI N42.22, and ANSI N13.30. These voluntary standards define a hierarchy of traceability with an unbroken linkage to NIST. Under the standards, the evaluation materials shall be composed of appropriate matrices (i.e., matrix categories commonly analyzed by the laboratory) and consist of appropriate (commonly encountered) activity concentration ranges. Current participants in the NRIP program requested these traceability evaluations as part of their quality assurance programs.

For more information see
http://physics.nist.gov/Divisions/Div846/Gp4/Environ/nrip.html.

Preliminary Agenda
Status - Participants and Matrix Selection
FY05 Results - Customary Program
FY05 Results - Emergency Preparedness Exercise
Participant's Comments and Suggestions
Group Discussions

Contact: Kenneth.inn@nist.gov

# Abstract: QA/QC implementation in real world gamma spectroscopy systems

**Luc De Baerdemaeker Ing. BSc MSc**
Director of Marketing
Radiochemistry Laboratory Products
CANBERRA

During recent years, quality control and quality assurance have gained increased attention from the operators of analytical equipment. The gamma spectrometer systems of today are not significantly different from those that have been employed for a number of years. However, the increased emphasis on quality assurance record keeping by regulators and radioanalysis laboratory customers and the improved database technology for storing large amounts of data that is available today have made it necessary and possible for effective QA software to be a key part of any count room. Electronic storage of the data is the easy part. Knowing what parameters to track, what types of QA tests to use and how interpret the QA results is more difficult and is critical to having a good QA/QC program. Since most operators of analytical instruments are audited on regular basis, a good QA record keeping package must make data readily-accessible and easily defensible.

Today, many kinds of QA database packages are available for the various types of analytical instrumentation. Each instrument has its own specific types of parameters that need to be monitored to maintain peak performance. Gamma spectroscopy systems are instruments designed to qualitatively and quantitatively measure the gamma emission from radioactive materials. Parameters that need to be monitored for these types of systems relate to the detector and its associated electronics.

Basically, gamma spectroscopy systems are designed to measure extremely low to relatively high quantities of radioactive material, and the key quality assurance task is to ensure that the instrument's performance is stable before, during and even after analysis of unknown samples. A good QA software package should ensure that the user knows if performance of the instrument has changed since the last efficiency calibration so that corrective action may be taken as soon as possible. As an example, measuring the same source in the same conditions should report the same radioactivity over time (accounting for decay). However, this single parameter approach has limitations and is insufficient to show what caused an eventual exception situation.

What we present here is additional parameters that should be monitored in order to maintain a gamma spectroscopy system at peak performance over the equipment's lifetime. Data from these parameters give early warnings as to possible problems with the system as well as information for troubleshooting the problems.

The information presented is based on years of intensive cooperation between many users of electronic QA packages and consultancy work performed by CANBERRA to help customers successfully pass their audits and achieve accreditations for their counting laboratories. The goal is to provide advice as to how to create an accurate and balanced QA program which can alert the user to any situation that may cause the system to report erroneous results.

**Abstract:** **Utilization of Ion Implanted Silicon Charged Particle Detectors in Low Background Alpha / Beta Counting Systems for Radon, Thoron and progeny compensation.**

**Bill Cross**
**Product Application Manager**
**Alpha / Beta Instruments**
**Canberra Industries, Inc.**

For well over thirty years, Gas Flow Proportional Counters have been the detector of choice for low background alpha / beta counting systems. While these detectors offer performance characteristics well suited for the analysis of air filters, swipes, evaporated water and other liquids and more, they are not without their drawbacks.

Gas flow proportional counters exhibit extremely low leakage currents and very low capacitance that make them ideal for the detection of alpha and low energy beta particles. While the noise of a typical gas flow proportional counting system is excellent, charge collection and thus charge recombination, is not.

Canberra Harwell initiated this endeavor with the development of the iCAM, intelligent Continuous Air Monitor. An algorithm was developed that evaluates a pulse-height spectrum for the presence of alpha peaks and/or beta continuum. If present, the three major Radon and Thoron peaks (8.78 MeV, 7.68 MeV and 6.0 MeV) are evaluated for their total contribution, particularly in the spectrum region where the peaks of Uranium, Plutonium, Americium, Curium, etc would be located. The contribution of these peaks is subtracted from the spectrum and the residual spectrum provides a clear indication of the presence or absence of the alpha peaks of these isotopes of concern.

Canberra Tennelec, with Canberra Harwell, extended the capabilities of this algorithm and has incorporated it into a Low Background Alpha / Beta Counting system for the analysis or air filters and swipes of nearly all types and sizes, the iSolo. The advantages of such a counting system are many. It does not, for example, require gas to operate and it dramatically reduces the time required to make an accurate decision about the condition of an area that may or may not have air borne, long half-life alpha emitters present. Information about the algorithm utilized for these analysis and the systems available that incorporate this approach to alpha / beta counting, will be presented.

# Eichrom Users' Group Workshop at the RRMC

# 51st Annual RRMC

Workshop organized by Eichrom Technologies, Inc.
Chairperson: Lawrence Jassin, jassin@eichrom.com

Abstract: Eichrom is a worldwide provider of products and services in the field of analytical chemistry, with offices in metro Chicago and Rennes, France. Registered to the ISO 9001:2000 standard, Eichrom focuses on developing analytical methodologies that set industry standards for measuring environmental contaminants such as radioactive elements, hazardous metals and dioxins.

This year's workshop will highlight research and initiatives from inside of Eichrom and by key scientists in the radiochemistry community. Sherrod Maxwell of Westinghouse Savannah River Company will present an update on a new, rapid actinide separation method that provides total dissolution of large soil samples, high chemical recoveries and effective removal of matrix interferences. This method uses stacked TEVA Resin ®, TRU Resin and DGA Resin cartridges that allows the rapid separation of plutonium (Pu), neptunium (Np), uranium (U), americium (Am) and curium (Cm) using a single multi-stage column combined with alpha spectrometry. The method combines a rapid fusion step to dissolve refractory analytes and matrix removal using cerium fluoride precipitation to process the difficult soil matrix. Bill Burnett of FSU will present radium analysis using $MnO_2$ Resin and automated counting. Details of this presentation are provided below.

The workshop will also serve as a forum to update our users' on new products, applications and the continuous improvement of our quality system. Details on beryllium analysis in very difficult matricies and new uptake curves for our Ln series of resins will be presented. We also plan to discuss manufacturing issues including the introduction of a new cartridge mold and the conversion to new non-ionic frits for our pre-packaged column line.

As part of our ISO 9001:2000 continuous improvement program, new quality control methods have been developed for our resins. These are based on at least two components separated with vacuum box assisted resin cartridges. A progress report will be provided on the implementation of the new procedures.

### Automated Radium Analyses Using $MnO_2$ Resin and a Radon Analyzer

Natasha Dimova, Henrieta Dulaiova, and Bill Burnett
Environmental Radioactivity Measurement Facility, Department of Oceanography, Florida State University, Tallahassee, FL 32306

Derek Lane-Smith, Durridge Co., Inc., 7 Railroad Avenue, Suite D, Bedford, MA 01730

Eichrom introduced $MnO_2$ Resin last year as a preconcentrator for radium isotopes in water. Sherrod Maxwell has developed a method based on $MnO_2$ Resin to concentrate radium from natural waters and then using Ln Resin and DGA Resin for purification and separation of Ac (for $^{228}$Ra). The alpha-emitting radium isotopes are assayed via alpha spectrometry after microprecipitation and mounting onto 0.1 $\mu$m filters.

We show here that one may also assay the alpha-emitting isotopes $^{226}$Ra and $^{224}$Ra using an automated approach that combines use of the $MnO_2$ Resin with a commercial radon-in-air analyzer (Durridge RAD-7). One simply passes a measured volume of a water sample through the resin that is loaded in a specially designed glass cartridge. The resin will quantitatively adsorb the radium up to flow rates of at least 100 mL/min. The cartridge is then connected to the radon analyzer via a simple system of tubes and valves and air is based through the resin that now emanates isotopes of radon ($^{222}$Rn and $^{220}$Rn) from their respective radium parents. The RAD-7 analyzer sorts these isotopes via energy discrimination. This initial analysis is done for the analysis of the short-lived $^{224}$Ra ($^{223}$Ra may also be detected via $^{219}$Rn if present in sufficient activities). After the first analysis, the stopcock on the glass cartridge is closed and the time noted for ingrowth of $^{222}$Rn from $^{226}$Ra. After about 4-5 days (~50% equilibrium), the cartridge is again plumbed into the RAD-7 for analysis. If one requires $^{228}$Ra as well, the $MnO_2$ Resin can be stripped with acid and Ac separated as in the Maxwell procedure.

# Calibrating Alpha/Beta Counters with Cs-137

"Instrument problems" reported to Protean following a failed cross-check or QA/QC analysis are sometimes caused by incorrect use of calibration standards. Different errors are reported, the most serious being under-reporting of sample activity. Common mistakes in the selection and use of calibration sources will be discussed, and examples of incorrect calibration source usage will be presented.

# The Eighth Annual Session on Quality Assurance and Quality Control:

# RRMC 2005
## Caesars Tahoe, Stateline, NV
## October 24 - 28, 2005

## A Forum for Radiochemistry: QC and QA in the Real World

**Objectives:** This session, held as part of the agenda of the 51st Radiobioassay and Radiochemical Measurements Conference (RRMC) offer you a time to present lessons learned in your lab, share practical tips, brainstorm ideas for improving quality control (QC) and quality assurance (QA), and learn about the major concerns of the QA/QC community in radiochemistry, radiobioassay, and related fields.

The workshop will offer as many topics as practicable, with presentations grouped by topic or area of interest. You are **invited and encouraged** to take part in this informal, maybe even fun, session, either by attending or by presenting information. In addition, if there are more ideas and papers than we can include in this session, we may be able to include an additional poster session on similar QC/QA ideas and issues.

**QA/QC in the Real World:** As the title suggests, this session is intended for the chemists, technicians, supervisors, quality assurance officers, data reviewers, and others who actually do the analytical work in the laboratory, or who work directly with data produced by the analytical labs. While lab directors, managers, statisticians, math whizzes, and all others interested are encouraged to attend and participate, we especially want to hear from those of you, "in the trenches" every day.

> What have you learned that will help others improve their data quality?
> What mistakes have you made, and how did you detect and correct them (and prevent them from happening again?)
> How does your lab handle validation of a new method?
> How good is good enough data?
> Do you have a spreadsheet or other software that helps you crunch the numbers better and faster?
> How do you use performance evaluation samples to improve data quality?
> Etc, etc, etc.

We want this session to reflect quality control and quality assurance in the real world!

**Draft Agenda Ideas:**

**Updates on Issues**

What's really going on with various programs, implementation projects, PE programs, accreditation, etc, and how will your laboratory or company be affected?

**Method Validation** (papers, ideas, overheads, provocative ideas are now being solicited for this session.)

When do you need to do it?
How good is good enough?
Do you need to hire a statistician?
How many replicates, duplicates, spikes, etc?
What statistical tests do you do?
Intralab vs. interlaboratory validation studies.
How can you tell when you are done and the method is acceptable?

**Data Evaluation (Verification)** papers, ideas, overheads, provocative ideas, are now being solicited for this session.)

Do you use data evaluation flags?
What do you look for in data verification besides contract compliance?
Do you use check lists, and can you share those?
How do you use batch QC in the verification process?
What does PE data tell you about the batch?
When do you reject data and reanalyze the sample?
If SOWs drive verification, do poorly written SOW lead to poor data?
If you do poorly on external and internal PE samples, do you continue to report client data?

**Uncertainty calculations, processes, questions, and answers.**

**Bits and Pieces, Words to the Wise, and Other Stuff** (papers, ideas, overheads, cartoons, slides, provocative ideas, weird solutions, and other "outside the box" ideas are being solicited for this session.

This is the place to come even if you have only a 5-minute presentation about a simple improvement you have made in your lab. This session promises to be fun and beneficial, with time allowed for questions, arguments, and alternative ideas.

If you have questions about any of the sessions, or would like to discuss an idea you have for presentation, please correspond by e-mail to Mary Wisdom at wisdom.mary@epa.gov. **Please submit a Note of Interest by September 9, to let us know that you hope to make a presentation.**

We will notify submitters by October 3, 2005, of abstracts accepted for presentations or posters.

DOE/NV/11718–1103 ABS

51st Annual Radiobioassay and Radiochemical Measurements Conference (RRMC)
Stateline, NV, October 24-28, 2005

Erik C. Nielsen*,Bechtel Nevada, Remote Sensing Laboratory-Nellis, P.O. Box 98521, M/S RSL-11, Las Vegas, NV 89193-8521

# *Radioanalytical Data Quality Objectives (DQOs) and Measurement Quality Objectives (MQOs) during a Federal Radiological Monitoring and Assessment Center (FRMAC) Response*

Oral Presentation

## ABSTRACT

During the emergency and intermediate phases of a nuclear or radiological incident that activates the National Response Plan (NRP), the FRMAC collects environmental samples that are analyzed by organizations with radioanalytical capability. During the emergency phase (Phase I) of an incident, analytical data will be urgently needed as a basis for protective actions. In the early stages of an emergency, the impact of the incident on the health and safety of the public is not well defined. Therefore, the resources dedicated to quality assurance (QA) activities must be sufficient to assure that appropriate radioanalytical Measurement Quality Objectives (MQOs) and assessment DQOs are met. As the emergency stabilizes, QA activities will evolve commensurate with the need to reach appropriate DQOs. The MQOs represent a compromise between precise analytical determinations and the timeliness necessary for emergency response activities. Results need to be "good enough" to make decisions appropriate to the level of risk associated with making an "incorrect" decision. Minimum Detectable Concentration (MDC), Lower Limit of Detection (LLD), and Critical Level Tests ($L_c$) can all serve as an expression of the MQO. Combined standard uncertainty and expanded uncertainty must be reported with each analytical result to allow decision makers to estimate the probability of making a decision error. The relationship between Protective Action Guides (PAGs), Derived Response Levels (DRLs), laboratory detection limits, and measurement uncertainties is described.

Keywords: FRMAC, health, safety

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# INTERCOMPARISON STUDIES SPIKED URINE SAMPLES USED FOR METHOD DEVELOPMENT AND VALIDATION

J. M. Rankin, G. F. Payne, and N.E. Bores
Intercomparison Studies Program
Quality Services Division
Oak Ridge National Laboratory*
Oak Ridge, Tennessee 37831

## Abstract

Oak Ridge National Laboratory's (ORNL) Intercomparison Studies Program (ISP) provides customers with natural urine samples spiked with known activity levels of radioisotopes. These samples are commonly used for quality control purposes in routine analytical operations. ISP also provides spiked urine samples for purposes such as method development and validation. Forty single-blind natural uranium spiked urine samples were supplied to ORNL's Radiobioassay Laboratory for use in the development of an improved method for the determination of $^{235}$U and $^{238}$U in urine by inductively coupled plasma-mass spectrometry. When the results from this sample set were compared to the actual spike activities, a small negative relative bias was found for thirty-nine of the samples. Based on their past performances and their confidence in the ISP spike sample preparations, the Radiobioassay Laboratory re-examined the method; and further changes were made to the procedure. The Radiobioassay Laboratory then analyzed an additional twenty uranium spiked urine samples using the modified procedure and found that the accuracy of the data set significantly improved.

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*Managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract DE-AC05-00OR22725.

# Automated Audit Software for Laboratory, Field Operations and QA Systems

Mo Khalil, Chemical Check Inc., and Richard Amano, Laboratory Data Consultants Inc.

An automated audit software program has been designed and developed to enhance the laboratory QA /QC program and improve the internal and external laboratory audit, and field sampling audits process and ease the process of tracking, findings, observations and corrective actions within the laboratory.

The Audit software program guides the auditor step-by step through DOEAP, and NELAC –style Audits. The software streamlines the audit preparation, onsite audit process, final reporting and long-term document storage and management.
The user builds the audit content in the master database and then downloads the audit to a "briefcase" database that contains the electronic DOECAP, and NELAC checklists and electronic versions of all pertinent reference documents including. Pdfs of past audits, internal audits, PT results, laboratory QA manuals, and other pertinent documentation.
The briefcase module is installed on a laptop, which the auditor will then take on-site to conduct the audit. At the conclusion of the audit the audit results, assessment report, corrective actions and all communications are up-loaded together in the electronic briefcase for easy archival retrieval and review.

The following are features of the new automated audit software:

- Wizard-style interface guide the auditor step-by step through the preparation for and execution of a DOECAP, and NELAC style audits.
- Reference documents are hyperlinked to the checklist for instant retrieval at anytime during the course of the audit.
- Automated generations of assessment reports and tabular summary of findings, observations and corrective actions.
- Tracks findings and corrective action response from the laboratory.
- Provide email notification for upcoming audits and critical dates in the audit process.
- Can be used for both internal systems audits and external laboratory audits as well as field sampling audit.

# DOE Lung Intercalibration Committee Workshop

The DOE Lung Intercalibration Committee Workshop will be meeting to discuss current topics and technical issues related to the in vivo measurement of radioactive material in workers. Particular attention is paid to the measurement of uranium, transuranic nuclides, and other radioactive materials that emit low energy photons. There are scheduled presentations and ad hoc presentations but time is also allotted for discussions of technical issues that arise from the information exchanges. The agenda is usually fluid to best meet the needs of the workshop attendees. Examples of just some of the topics that could be discussed include methods for quantitative measurement of uranium and plutonium in the lungs and other organs, the DOE Laboratory Accreditation Program, operational issues, measurement quality control, instrumentation, calibration, and calibration phantoms.

# 51st Annual Radiobioassay & Radiochemical Measurements Conference Oral Presentation Abstracts

# Technical Program Schedule - Oral Presentations

**Wednesday, October 26, 2005 - Keynote Speaker**
**Chairperson: Carolyn Wong**

| | | | |
|---|---|---|---|
| 8:20 AM | Darleane Hoffman | University of California, Berkeley | The Crisis in Radiochemistry and Nuclear Chemistry Education |
| 9:00 AM | Mark Hart | University of California, Lawrence Livermore National Laboratory | Radiation - What is Important? An Exhibit of Exempt Radioactive Items |
| 9:25 AM | Virgene IdekerMulligan and Bob Shannon | Kaiser Analytical Management Services, Inc. | They Said It Couldn't Be Done… |

**Wednesday, October 26, 2005 - Standards & Standards Testing / QA / QC**
**Chairpersons: Bob Shannon & Matt Lardy**

| | | | |
|---|---|---|---|
| 10:10 AM | Donna M. Beals, David E. McCurdy, and Daniel E. Montgomery | Savannah River National Lab | Opportunities for the Advancement of Science Through Participation in the ASTM International |
| 10:35 AM | James Dahlgran, Mary Verwolf, David Sill, James Filliben, Gavin Spalletta, Kenneth G.W. Inn | RESL; NIST | Draft PT Uncertainty Acceptance Criteria Derived from Historical MAPEP Results |
| 11:00 AM | Cheryl Antonio | Pacific Northwest National Laboratory | Lessons Learned: Incidents Can Bring Unwelcome Surprises |
| 11:25 AM | Natasha Dimova, Bill Burnett and Henrieta Dulaiova | Environmental Radioactivity Measurement Facility, Department of Oceanography, Florida State University | Preparations of Radium Standards for Calibration of a Delayed Coincidence Counter System |
| 11:40 AM | T. L. Rucker | Science Applications International Corporation | Application of Decision Levels (DLs) and Minimum Detectable Amounts (MDAs) – Will the Madness Ever End? |

# Technical Program Schedule - Oral Presentations

**Wednesday, October 26, 2005 - Mass Spectrometry**
**Chairpersons: Dave McCurdy & Cindy Conrado**

| | | | |
|---|---|---|---|
| 1:20 PM | S. S. Kannamkumarath, G. R. Rao, B. D. Gonzalez, R. D. Myers, D. K. Hatmaker, and M. Thein | Oak Ridge Institute for Science and Education; Operational Safety Services Division Oak Ridge National Laboratory | An Improved Method for the Determination of $^{235}U$ and $^{238}U$ in Urine-By Inductively Coupled Plasma-Mass Spectrometry |
| 1:45 PM | Julian Wills, Meike Hamester, Chuck Douthitt | Thermo Electron | Application of extended dynamic range Sector Field ICP-MS to the analysis of high uranium matrices |
| 2:10 PM | Dominic Larivière, Stephen Kiser, Chunsheng Li, Vladimir Epov, R. Jack Cornett | Radiation Protection Bureau, Health Canada; Trent University | New and fast analytical protocol for the determination of actinides in air filter samples by HPLC-ICP-MS |
| 2:35 PM | Bogen, K.T., T.F. Hamilton, T.A. Brown, A.A. Marchetti, and R.E. Marinelli | University of California, Lawrence Livermore National Laboratory | Age-related trend in elevated plutonium-239 measured by AMS in urine samples collected in 1998-2003 from Enewetak residents and Rongelap resettlement workers |

# Technical Program Schedule - Oral Presentations

**Thursday, October 27, 2005 - Bioassay: In Vitro**
**Chairpersons: Gary Kramer & Sandi Fisher**

| | | | |
|---|---|---|---|
| 8:00 AM | Evan Crawford and Jerry LaRosa | Yale University; NIST | What a Coincidence-Developing a Rapid LSC Screening Method for $^{90}$Sr Contamination in Urine |
| 8:25 AM | John S. Morton, Robert P. Wills, Robert D. Timm, James B. Westmoreland | General Engineering Labs | On-site Collection, Concentration and Separation of Nuclide Specific Analytes in Urine |
| 8:50 AM | D. J. Fauth and T. R. La Bone | Westinghouse Savannah River Company | Validation Process for Bioassay MDCs |
| 9:15 AM | Bogen, K.T., D.P. Hickman, T.F. Hamilton, T.A. Brown, C.C. Cox, A.A. Marchetti and R.E. Martinelli | University of California, Lawrence Livermore National Laboratory | AMS Analysis of $^{239}$Pu in Archived Occupational Samples |

**Thursday, October 27, 2005 - Bioassay: In Vivo**
**Chairpersons: Stan Morton & Ilham AlMahamid**

| | | | |
|---|---|---|---|
| 10:00 AM | Gary H. Kramer and Barry M. Hauck | Human Monitoring Laboratory, Radiation Protection Bureau, Health Canada | The Human Monitoring Laboratory's new whole body counter, its comparison with the previous system and an extension of the calibration using Monte Carlo simulations |
| 10:25 AM | Matthew Mille and Svetlana Nour | University of Maryland; NIST | Getting into the BOMAB Head |
| 10:50 AM | Rachel A. Zeman, Sandra K. Fisher, David P. Hickman | University of California, Lawrence Livermore National Laboratory | The Heart Must be Removed Prior to In Vivo Lung Counting to Achieve Optimum Detection Efficiency of Low Energy Gammas |
| 11:15 AM | Malcolm Ennis, Andy Baker and Doug Van Cleef | Los Alamos National Laboratory; Advanced Measurement Technology, Inc | High Resolution HPGe Lung Counting Without Liquid Nitrogen |
| 11:40 AM | Gary H. Kramer and Barry M. Hauck | Human Monitoring Laboratory, Radiation Protection Bureau, Health Canada | The sliced BOMAB phantom: a new variant for intercomparisons. |

# Technical Program Schedule - Oral Presentations

**Thursday, October 27, 2005 - Environmental Measurements**
**Chairpersons: Bahman Parsa & Shiyamalie Ruberu**

| | | | |
|---|---|---|---|
| 1:20 PM | Mark A. Verdoorn, Dennis W. McBride | Idaho National Laboratory | Development of Autonomous Unmanned Vehicles for Radiation Surveys and Sampling |
| 1:45 PM | Abdul Khalique, Richard Pietz, and Richard Lanyon | Metropolitan Water Reclamation District of Greater Chicago | Radon Monitoring at the Metropolitan Water Reclamation District of Greater Chicago |
| 2:10 PM | Sherrod L. Maxwell | Westinghouse Savannah River Co. | Rapid Emergency Method for Actinides and Sr-89/90 in Water Samples |
| 2:35 PM | Tosheva Z.P., Kies A.F., Hofmann | University of Luxembourg | Investigation of radioactivity in waters in Luxemburg by follow-up radiochemical method |

# Technical Program Schedule - Oral Presentations

**Friday, October 28, 2005 - Radiometric Methods & Procedures**
**Chairpersons: Barry Stewart & Donna Beals**

| | | | |
|---|---|---|---|
| 8:00 AM | Cecilia C. DiPrete, Timothy A. DeVol, Christopher D. Theisen, and David P. DiPrete | Savannah River National Laboratory; Clemson University | Effect of Quench on Alpha/Beta Pulse Shape Discrimination of Liquid Scintillation Cocktails |
| 8:25 AM | Hiromu Kurosaki, Payam Motabar, and Ken Inn | National Institute of Standards and Technology; University of Maryland | Magic or Science? ~$^{90}$Sr efficiency calibration data reduction |
| 8:50 AM | J. La Rosa, I. Outola, E. Crawford | Ionizing Radiation Division, National Institute of Standards and Technology | Verification of $^{237}$Np activity concentration in a mixed radionuclides solution: recent experiences in chemical separation and measurement by alpha spectrometry |
| 9:15 AM | Shiyamalie R. Ruberu, Yun-Gang Liu, and S. Kusum Perera | California Department of Health Services | Occurrence and Distribution of $^{210}$Lead and $^{210}$Polonium in California Groundwater Determined by a Single Sample Analysis Method |

**Friday, October 28, 2005 - Radiometric Methods & Procedures**
**Chairpersons: Ken Inn & Ann Mullins**

| | | | |
|---|---|---|---|
| 10:00 AM | Payam Motabar, Svetlana Nour and Kenneth G. W. Inn | National Institute of Standards and Technology | Effect of Energy on Alpha-particle Counting Efficiency |
| 10:25 AM | M. Sadeghi, P. Van den Winkel, H. Afarideh and M. Haji-Saeid, L. De Vis, R. Waegeneer,A. De Schriiver. | Islamic Azad University; Center for Agriculture & Medicine (NRCAM); VUB-Cyclotron | Centrifugal electrodissolution system for irradiated rhodium target material used for the cyclotron $^{103}$Pd production |
| 10:50 AM | Barry Stewart | Carlsbad Environmental Monitoring and Research Center | Use of Thin Films to Control Recoil Contamination of Alpha Spectrometry Detectors |
| 11:15 AM | Edward Hall and Kenneth G.W. Inn | University of Rochester; NIST | Massive Impact. The Use of Collodion Films to Shield Solid State Alpha Detectors from Heavy Ion Recoils |
| 11:40 AM | Steven Wheland and Matt Lardy | STL Richland | Evaluation of Quench Corrected Liquid Scintillation Counting Instrument Blanks to Reduce False Positive Rate |

# Technical Program Schedule - Oral Presentations

**Friday, October 28, 2005 - Keynote Speaker**
**Chairperson: Bryan Bandong**

| | | | |
|---|---|---|---|
| 1:00 PM | Ken Moody | University of California, Lawrence Livermore National Laboratory | Discovery of Superheavy Elements |

**Friday, October 28, 2005 - Homeland Security / Emergency Response**
**Chairpersons: Bryan Bandong & Rod Melgard**

| | | | |
|---|---|---|---|
| 2:55 PM | Joel Swanson | University of California, Lawrence Livermore National Laboratory | The Radiological Assistance Program |
| 3:20 PM | Donna Beals and Doug Van Cleef | Savannah River National Lab; ORTEC | Using Existing Field Radiological Instrumentation to Influence Emergency Response Decisions |
| 3:45 PM | Gary H. Kramer | Human Monitoring Laboratory, Radiation Protection Bureau, Health Canada | Anatomy of an emergency exercise: EXFO. |
| 4:10 PM | Iisa Outola, Svetlana Nour, Hiromu Kurosaki, Jerry LaRosa, Kenneth G.W. Inn and Lynn Albin | NIST, MS 8462; Washington State Department of Public Health | Improvement in Radioassay Emergency Response Capability: NRIP'05 |
| 4:35 PM | Carolyn Wong, Robb Hadley, Mark Johnson, Dann Haynes | University of California, Lawrence Livermore National Laboratory | The Livermore HOTSPOT Mobile Laboratory |

# Radiation - What Is Important?

## An Exhibit of Exempt Radioactive Items

Over 101 radioactive items, including antiques, contemporary items, consumer products, gems, minerals, and fossils will be on display. Many people viewing the exhibit have commented upon seeing similar items in their home or their grandparent's home. This display offers a unique look at an extensive collection of objects that lend an informative perspective on naturally occurring radiation in the environment and its everyday presence in our lives. Examples of items that will be on display include glass objects that glow green, the rare "Lone Ranger Atomic Bomb Ring", the historic "Revigator", a colossal specimen of pitchblende ore weighing two pounds, a radioactive brontosaurus rib bone, and many, many, others.

Jane and Mark Hart have used this exhibit as an integral part of over 100 presentations they have given to professional and public audiences, and news media during the past 11 years. Pieces in their personal collection of over 500 items were found in antique shops, art stores, flea markets, eBay, and even hardware and grocery stores. Items in their collection originate from the United States, Austria, Czech Republic, Russia, Zaire, Germany, Switzerland, Canada, Brazil, India, and Burma. Their speaking engagements have included the 42nd Annual Radiobioassay & Radiochemical Measurements Conference, San Francisco; Stanford University; U.C. Davis; Westmont College, Santa Barbara; U.S. Military Academy, West Point; Naval Postgraduate School, Monterey; Scientific Delegation from Doha, Qatar; Sandia National Laboratories; Lawrence Livermore National Laboratory; Savannah River Laboratory; United States Coast Guard; Health Physics Society; Canadian Radiation Protection Association; American Nuclear Society; American Chemical Society; Industrial Hygiene Society; San Francisco Gem and Mineral Society, Pacific Grove Museum of Natural History, high schools, middle schools, and church groups.

# They Said It Couldn't Be Done . . .

Virgene IdekerMulligan and Bob Shannon
*Kaiser Analytical Management Services, Inc.*

For more than three decades Rocky Flats was a high-security, top-secret factory that made plutonium triggers for the United States Government's nuclear arsenal. Today, there are few remaining visual cues to the history of Rocky Flats. The site -- at 6,266 acres, nearly half the size of Manhattan -- is being converted into a wildlife refuge on the northwestern edge of the Metropolitan Denver Area. On the site of the building once known as the most dangerous in America, B771, where liquid plutonium often leaked from faulty pipes and valves, there is only a large patch of replanted land covered by coconut matting to prevent erosion.

1995 estimates put the cost for closure at $22-36 billion and the completion date around 2060. In 1995 Kaiser-Hill committed to cleaning up and closing down the Site by the end of 2006 for less than $7 billion

Decontaminating, demolishing and disposing of Rocky Flats - in the end, a $3.96 billion task slated to be finished in October 2006 - has involved a series of ''unbelievable'' technological feats:

- *800 structures have been demolished;*
- *More than 39,500 containers and 20,000 cubic yards of highly radioactive transuranic waste have gone to their final resting place at the Waste Isolation Pilot Plant in Carlshad, N.M;*
- *21 tons of weapons-grade nuclear materials and 30,000 liters of plutonium and enriched uranium solutions have been stabilized, and the material packed and dispositioned;*
- *425,000 $m^3$ of low-level radioactive and mixed waste have been shipped to disposal sites.*
- *Environmental contamination in the soil and groundwater, old landfills, contaminated sites have been characterized and addressed. All of this had to be accomplished while ensuring worker and general public safety.*

Before buildings could come down, they needed to be decontaminated. This included decommissioning/decontaminating, removing and disposing of massive pieces of highly toxic and radioactive equipment requiring new methods for decontaminating and disposing of highly contaminated equipment. Landfills and contaminated areas needed to be remediated.

Each of these tasks required dependable and defensible analytical data to guide its progress and to demonstrate that goals had been reached. Given the accelerated pace of the closure activities, there was an overwhelming volume of testing (environmental, waste, industrial health and safety and radiobioassay) needed to be accomplished with ever-increasing urgency. It was immediately clear (not only to the skeptical) that thinking outside the box would be the only way to meet this seemingly unachievable goal.

This presentation will address key event pictures and numbers and will discuss many of the analytical challenges and realities, as well as engineering and analytical management innovations, which facilitated bringing the project to closure ahead-of-schedule and under-budget.

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# Opportunities for the Advancement of Science Through Participation in the ASTM International

Donna M. Beals[1], David E. McCurdy[2], Daniel E. Montgomery[3]

ASTM International (formerly the American Society for Testing and Materials) was established in 1898 to write and develop consensus standards for use in government and industry. It consists of 180 technical committees, with members from industry, government, private and university sectors. The approval of a new Test Method for publication by ASTM International begins with the submittal of a method by a member based on an industry need or government regulation. A single operator test (SOT) plan is prepared, the SOT conducted, and the results reviewed at the Subcommittee level. Upon successful completion of the SOT the Subcommittee may prepare an inter-laboratory (round-robin) test plan with the approval of an ASTM Results Advisor. At this point analytical laboratories are solicited for participation in the study. The participating lab is provided with the proposed Test Method, calibration standards and samples to be tested. A successful study requires that at least six laboratories return acceptable results. These results are then evaluated to prepare the precision and bias statement that is included with the final published Test Method.

The benefit to the participating laboratories is the opportunity to test a new, proven analytical method for an analyte of interest without having to expend the development time and money often required for new procedures. Prior to the round robin testing the method has been tested and reviewed for robustness and applicability by a qualified Subcommittee, such that no additional research should be required. The requirements placed upon the participating laboratories include that they follow the Test Method as written and report the results back to the Subcommittee in a timely fashion.

This presentation will include an example of a recent Test Method development and validation by round-robin testing. A comparison of the ASTM International validation process to the US EPA process, as it applies to radionuclides in drinking water, will be presented. The presentation will provide the audience with a listing and status of radiochemical methods within the ASTM International currently under development for which round-robin testing will be required, and the contact information for interested laboratory personnel.

## Test Method Development and Validation

The ASTM has recently published a new Standard Method, D7168, "Standard Test Method for the Determination of Technetium-99 in Water by Solid Phase Extraction." This Method was sponsored by Subcommittee D19.04 on Radioactivity in Water. The process actually began in 1997 when the 3M Co. and Eichrom Industries co-sponsored a proposed new standard on the analysis of Tc-99 in water using each company's solid

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phase extraction disk. The first single operator tests (SOT) were performed using a Tc-95m yield monitor and reported to the Subcommittee at the ASTM D19.04 meeting held in conjunction with the 1998 BAER Conference. The results were not acceptable. In June 2000 another set of SOT data was presented using the spiked duplicate method to determine chemical yield. These results were acceptable and the Subcommittee began the process of writing the final method and drafting the inter-laboratory study.

The Testing Plan for the inter-laboratory study was approved by the ASTM Results Advisor in 2001 and the samples submitted to the participating labs in 2002. Fifteen labs received samples, the Method to be used, all the solid phase extraction disks required, a filter flask, and enough NIST traceable Tc-99 to perform all the required calibration checks and analyses. The DOE Radiological and Environmental Services laboratory prepared the test samples and calibration standards for distribution. Twelve labs returned results to the Subcommittee by the end of the year. The returned data was evaluated with respect to ASTM D2777, "Practice for the Determination of Precision and Bias of Applicable Test Methods of Committee D19 on Water." A summary of this data was presented at the 2003 RRMC. The precision and bias statements for inclusion in the Test Method were approved by the Results Advisor in 2004 and the Test Method began the ballot process, receiving Society approval and publication in 2005.

Of the twelve labs that submitted sample results, two labs appeared to have difficulty with variation A and one lab with variation B such that the data was excluded from the final statistical analysis (variation A and B were the use of the Eichrom disk or the 3M disk). The lab that seemed to have difficulty with variation B was not one of the labs that had difficulty with variation A. None of the participating labs indicated any significant problems with the Method. Feedback has been provided to the participating labs in the case that they may want to continue using the new Method. Now that it has been published by ASTM the participating labs may use the Method and claim ASTM pedigree for their analyses.

**ASTM vs. EPA Certification**

The US EPA Office of Water has responsibility for approving analytical methods for use with drinking water. Although Tc-99 is not a priority pollutant required to be separately monitored in drinking water, the ASTM validation process discussed above can be used for comparison to the EPA Drinking Water Certification process. Both processes begin when a need is identified to measure an analyte in water due to a government regulation (and/or industry need for ASTM). The ASTM process begins with a member proposing the new method and then a SOT is conducted resulting in data for review and approval. This is followed by an inter-laboratory study, which must be approved by the ASTM Results Advisor prior to initiating. At least six labs must return acceptable results for the study to be valid. The data is reviewed by the Subcommittee and the ASTM Results Advisor and then the Method is submitted for ballot approval by the Committee and Society as a whole. Publication in the ASTM Annual Book of Standards is the final step.

The EPA process begins with a laboratory submitting a formal application to the EPA to allow the use of an alternative test method, including a study plan that demonstrates equivalence with the existing EPA method (Alternative Test Method Approval Process). The proposing lab must conduct the study and submit the results to the EPA for evaluation and validation. If accepted the EPA can begin the rule making process to publish the new method in the Federal Register.

A comparison of the number and types of samples that must be completed for ASTM and EPA approval is shown in Table 1. As seen in Table 1, the ASTM requires much more testing at the SOT level, as this is the method development. Variable sample volumes, matrices (or added interferences) and spike levels are tested by the method sponsor or during the SOT. The only prescribed value in the SOT comes from D5847, "Practice for Writing Quality Control Specifications for Standard Test Methods for Water Analysis," which requires 7 blanks to be run prior to performing a new method. Similar development studies would be documented in the application to the EPA however there are no defined guidelines as to the amount of data required.

**Table 1. Number of Analyses Required for Method Approval**

| organization | # samples | # matrices | # volume | # spk level | # for blank | # for MDL | | | total # analyses |
|---|---|---|---|---|---|---|---|---|---|
| **Single Operator Testing** | | | | | | | | | |
| ASTM | 3-5 | ~3 | ~3 | ~3 | 7 | | | | ~100 |
| EPA | | | | | | | | | |
| **Interlaboratory Testing** | | | | | | | # of labs | # for P&B | |
| ASTM | 6 | 1 | 1 | 3 | 1 | | >6 | >36 | >42 |
| EPA | 3 | 3 | 1 | 3 | 5 | 7 | 3 | 12 | 84 |

For the inter-laboratory testing the ASTM requires at least six labs submit acceptable results. Each results package consists of a blank plus six samples (spiked at three levels) of a single matrix for a total of at least 42 results. For the EPA, only three labs are required to participate. However, each lab must perform a background characterization (5 blanks), a MDL study (7 samples), and a ruggedness study. The ruggedness study consists of three distinct samples, run straight, spiked and as a spike duplicate (total of 9 samples) plus 7 additional QC samples. This is a total of 28 samples per lab for a total of 84 results.

## ASTM Methods Currently Under Development

Several new Test Methods are currently in various stages of development in the D19.04, Radioactivity in Water, Subcommittee. These are listed in Table 2, along with the Task Group Chair who is responsible for the Method. Two methods are shown in Table 2 as requesting volunteers for inter-laboratory (IL) participation. The current priority is to complete the Pb-210 in water method first. Participating labs will be provided with the analysis method and prepared samples for analysis. The samples will be spiked at three different levels, two samples at each level. The Pb-210 tracer required for calibration will be provided to the lab, as will sufficient extraction chromatography columns to complete the study.

The method has gone through several SOTs and is now considered ready for testing by labs not previously familiar with the technique. Any lab that anticipates future work, or that is currently performing Pb-210 analyses but would like to evaluate a new, potentially more robust method, is welcome to participate in the inter-laboratory study. In this way the participating lab is provided a new analysis method that may lead to future work. The laboratory is providing an invaluable service to the radiochemistry community by helping to determine the precision and bias of a new method so that data by various methods can be compared to solve environmental issues.

**Table 2. Methods under Development in ASTM D19.04**

| **Analyte** | **Analytical Method** | **Counting Method** | **Task Group Chair** | **Stage of Development** |
|---|---|---|---|---|
| Gross α/β in water | Evaporation | LSC | Donivan Porterfield | Requesting IL participants |
| Sr-89 in water | Extraction chromatography | Cherenkov (LSC) | Donna Beals | Completing SOT |
| Sr-90 in vegetation | Solid phase extraction | LSC or gas filled detector | Doug Van Cleef | (Guide) need SOT |
| Pb-210 in water | Extraction chromatography | Proportional counting | Ann Mullin-Cleveland | Requesting IL participants |
| Alpha emitting Ra isotopes in water | Extraction chromatography | Alpha spectrometry | Ann Mullin-Cleveland | Completing SOT |

# Draft PT Uncertainty Acceptance Criteria Derived from Historical MAPEP Results

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The NELAP Performance Testing subcommittee on Radiochemistry has been considering the application of ANSI N42.22 test criteria as an indication of acceptable radioanalytical capabilities. The ANSI N42.22 test criteria asks that the difference between the laboratory result and the reference value (supported by measurement traceability links to NIST) be less than three times the combined standard uncertainty of that from the laboratory and the reference laboratory. This effort will provide the users of the PT exercises with a quantitative link between measurement results and the confidence in the results. One potential problem is that the laboratory could have an unusually high probability of passing the PTs when its reported uncertainty is larger than expected.

The NELAP PT Radiochemistry subcommittee has been evaluating establishing upper bounds to the uncertainties reported by the laboratories based on recent historical results being collected by the PT Provider for a limited number of drinking water gamma-ray emitting radionuclides, radium isotopes, uranium, and $^{90}$Sr. For other radionuclides in additional matrices to water, the DOE/RESL Multi-Analyte Performance Evaluation Program (MAPEP) has historical data that was mined in this study to draft additional bounds of uncertainties.

While individual studies indicated that the percent difference from the reference value was best characterized by symmetrical distributions, there was study to study fluctuations in distribution location. The bootstrap method was used on the residuals to characterize the 99 percent confidence level uncertainty function for each radionuclide in each matrix. The derived uncertainty limits are compared to historical results to verify appropriateness.

## Lesson Learned: Incidents Can Bring Unwelcome Surprises

Cheryl Antonio

In December 2004, the Internal Dosimetry Program (IDP) for the U.S. Department of Energy Hanford Site submitted two fecal samples for isotopic plutonium and americium-241analyses. Unbeknownst to IDP, the Hanford Site Contractor, and to the lab, those fecal samples had extraordinarily high levels of plutonium-239+240, in excess of 100,000 dpm. The fecal samples were collected as part of an incident follow-up. Based on preliminary indicators from the field and chest counts obtained from the workers, the plutonium-239+240 activity in the fecal samples was expected to be elevated, but not in excess of 1,000 dpm. Therefore the analytical laboratory was not notified of a potentially high activity bioassay sample.

Upon analysis, the first fecal sample from worker A was found to be of such high activity of plutonium that a valid measurement result could not be obtained. As a result of a preliminary investigation, all samples processed in the same batch as worker A's sample as well as those analyzed in the following batch were determined to be contaminated. A second analysis of Worker A's fecal sample determined that the activity was approximately 113,000 dpm for plutonium-239+240 and 3,500 dpm for americium-241. The cross contamination posed a serious problem during sample digestion because the bioassay samples processed in the same batch as Worker's A could not be replaced. The resulting laboratory contamination delayed the analysis of 24 incident response fecal samples for approximately 2 weeks while the lab decontaminated and replaced equipment.

Two important lessons learned were the importance of prompt notification to the lab for potentially elevated bioassay samples, and the value of a precautionary screening of incident response bioassay samples.

# PREPARATIONS OF RADIUM STANDARDS FOR CALIBRATION OF A DELAYED COINCIDENCE COUNTER SYSTEM

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We use a delayed coincidence counter (DCC) to measure low level Ra-224 and Ra-223 in coastal waters. This method was initially developed by Moore and Arnold (1996). Radium isotopes adsorb quantitatively on $MnO_2$ coated fibers ("Mn fibers") and their short-lived radon daughters (Rn-220 and Rn-219) are counted on the system. The system is calibrated using Th-232 and Ac-227 adsorbed onto a Mn fiber. In the process of standard preparation we found that thorium and actinium are not always retained quantitatively on these fibers as believed before. Alpha spectrometry analysis of the effluent showed that a significant amount (about 40 to 50%) breaks through when pure solutions are used. There is less break through when buffered solutions (as seawater) are used. We have investigated different parameters (pH, tracer source and media composition) to find the optimum conditions for the thorium and actinium adsorption.

# Application of Decision Levels (DLs) and Minimum Detectable Amounts (MDAs) – Will the Madness Ever End?

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## Introduction

The concept of minimum detectable amount (MDA) has long been established.[1] Lloyd Currie[2] helped to bring a standard definition to the detection limit concept and to develop a consistent statistical approach to the determination of limits. Currie gave it the symbol $L_d$ and defined it as the smallest quantity of radioactive material that can be detected (distinguished from background) with some specified degree of confidence. The MDA is the smallest amount (activity or mass) of an analyte in a sample that will be detected with a probability $\beta$ of non-detection (Type II error) while accepting a probability $\alpha$ of erroneously deciding that a positive (non-zero) quantity of analyte is present in an appropriate blank sample (Type I error). If expressed as a concentration, it is usually referred to as the minimum detectable concentration (MDC). The MDA is useful *a priori* in comparing different method's measurement capabilities and ability to show compliance with regulatory limits. Currie also developed an associated concept, the critical level ($L_c$) [also know as decision level, (DL) or critical value (MARLAP)], which is defined as the net signal or result that must be exceeded before there is a specific degree of confidence that the sample contains radioactive material (above background or that of the blank). The DL is the value that is used *a posteriori* for determining the presence of radioactivity in a sample by accepting a probability $\alpha$ of erroneously deciding that a positive (non-zero) quantity of analyte is present in an appropriate blank sample (Type I error). These two concepts are defined in relation to the measurement uncertainties as shown in the equations and illustrated in Figure 1 below:

$$MDA = k_\alpha \sigma_0 + k_\beta \sigma_d$$

and

$$DL = k_\alpha \sigma_0$$

where $k_\alpha$ and $k_\beta$ are the abscissas of the standardized normal distribution for the blank and MDA distributions, respectively, at the chosen probabilities (e.g., 1.65 each for 95%), $\sigma_0$ is the standard deviation of the net measurement result when the sample contains zero radioactivity, and $\sigma_d$ is the standard deviation of the net measurement result when the sample contains radioactivity at the level of the MDA. Many others[3,4] have elaborated on Currie's definitive work, and today his concepts are generally regarded as the standard approach for radiochemical analyses.

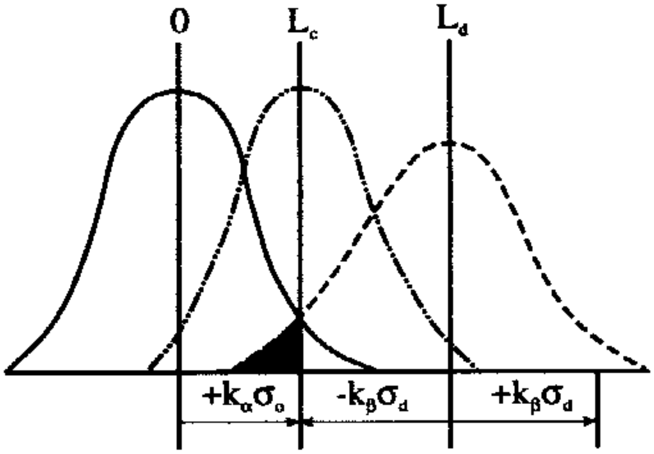

**Figure 1. DL ($L_c$) and MDA ($L_d$)**

## Problems

These concepts have been captured in National Standards (e.g. ANSI N13.30 and ANSI N42.2) and Regulatory Guidance Documents (e.g. MARLAP) and have been discussed numerous times in workshops at this conference. However, there continues to be much confusion over the application of decision levels (DLs) and minimum detectable amounts (MDAs) by both commercial laboratories, data validators, and many Department of Energy project managers. Many laboratories and projects are misapplying the MDA by making detection decisions based on comparison to it rather than the DL. One DOECAP Auditor has estimated that more DOE sites are using MDA for detection decisions than are using the DL. This practice likely stems from a confusion of the MDA as reported by most radioanalytical laboratories with the method detection limit (MDL) as defined by EPA for chemical analyses. However, MDLs equate to DLs in practice as they are both calculated as the upper confidence limit on a blank population and are to be used for making detection decisions.

The definition of the MDA presupposes that an appropriate detection threshold (i.e., the decision level) has already been defined. The $\alpha$ probabilities for the MDA and the DL are by necessity the same. If one uses the MDA for making detection decisions, they may be setting a low false positive rate but are setting up a very high false negative error rate for samples whose true concentration is as much as the MDA. That is, a 50% false negative error rate (one of every two samples) that have the analyte in them at the MDA level will be stated to be non-detects using that practice. Therefore, the true MDA (e.g., the level at which you have 5% false negatives) is now higher and the quoted MDA is no longer applicable.

In addition, practical application of Currie's concepts to radiochemical analyses have taken many different paths and have been combined with various assumptions, resulting in confusion on the part of many as to their calculation and use. These assumptions have been made in order to simplify the application of these concepts to practical measurements, and they generally relate to the determination of the standard deviation of the measurements. The typical use of the Currie equations has generally included only the counting uncertainty in the standard deviation of the measurement, since it is easily estimated (by taking the square root of the background counts and assuming paired observations). This has led some to believe that the DL should only make distinction from the detector background. However, the counting uncertainty may be only a portion of the total random uncertainty for many radiological analyses that involve sample preparation and chemical separation steps. Distinction from blanks, not detector background, should be desired. Furthermore, backgrounds are often well defined rather than made as paired observations.

Traditional formulas that have been presented in guidance documents and standards for the calculation of radiological MDAs and DLs have often been misunderstood and misapplied due to a lack of understanding of the assumptions they are based on.[5] Their application has also been confused since laboratories have usually been contractually required to calculate and report the MDA or both of them for each sample. Although this may be appropriate for DLs, it is not appropriate for MDAs which are intended to be *a priori*. This coupled with confusion over how to incorporate all of the uncertainty into the calculations has led to frustration on the part of laboratories and data users alike. Furthermore, many have worried about the fact that expected false positive (type I error) rates are not truly achieved by the equations due the discrete nature of the Poisson distribution, especially at low numbers of counts such as in alpha spectroscopy.[6,7]

## Solutions

We have previously recommended an approach for determining MDAs based on Currie=s basic definition but using data from populations of blanks for determination of the standard deviation used in the equations and replacing $k$ with the Student=s $t$ factor for the appropriate number of degrees of freedom.[8] The equation for the MDA becomes:

$$MDA = t^2 + 2\,ts_0$$

where $s_0$ is the standard deviation of a number of net blank results, and $t$ is the student's $t$ factor for the number of blank samples used to determine $s_0$ and for the chosen probabilities. This approach accounts for all uncertainty in the measurements, not just counting uncertainty. Using a population of method blank results to calculate $\sigma_{blank}$ is appropriate for calculation of MDAs since MDA is an *a priori* concept and not intended to be sample specific. This approach has proven to work well for defining sensitivity requirements for contract laboratory work and for other *a priori* applications.

We have also recommended that a similar approach can be used for determining the DL.[9] When the detector background or appropriate blank information is available, the critical level may be estimated by the following equation:

$$DL = \frac{t \bullet s_0}{E \bullet R \bullet IDF \bullet W}$$

where the DL is in units of disintegrations/minute (dpm) / unit sample, $s_0$ is the standard deviation of a set of appropriate blank net count rates (counts/minute) after detector background subtraction for blanks counted for the same length of time as the sample, $t$ is the student $t$ factor for appropriate number of degrees of freedom for the blank set, $E$ is the fractional detector efficiency (counts/disintegration) for the sample, $R$ is the fractional chemical recovery for the sample, $IDF$ is the ingrowth or decay factor for the sample, and $W$ is unit weight or volume of sample. This equation is a simple adaptation of Equation (1) by estimating $\sigma_0$ from a discrete number of blanks and using the sample specific parameters to come up with DL in units of dpm/unit sample. The $s_0$ may be calculated in terms of counts if desired if it is then divided by the count time. Either way, however, the assumption is made that the count times and background counts (and, therefore, the counting uncertainty) are the same or similar for all samples and blanks. Therefore, effort should be made to keep these as constant as possible. This may be difficult when multiple detector systems are involved for the analysis.

There is another approach, however, that is even easier for all involved and can yield an even more sample specific decision. A detection decision can be made using a DL estimated by the following equation:

$$DL = k_\alpha \bullet TPU_R$$

where the DL is in dpm/unit sample, $k_\alpha$ is the abscissa of the standardized normal distribution at the chosen probability (1.65 for 95%), $TPU_R$ is the total propagated uncertainty of the result (1$\sigma$ dpm/unit sample). This equation provides a DL that is almost equivalent to the DL calculated from Currie=s equation for sample results close to or less than the DL. The only difference in the two equations/approaches is the relatively small difference in the standard deviation at the DL level count rate value versus at the blank level. Even though the total propagated uncertainty (TPU) will be larger for samples larger than the DL, this equation can always be used for the detection decision since the sample result value will increase at a faster rate than the TPU value. Using this equation actually evaluates the hypothesis that the true result is greater than zero at the chosen probability level. Although this is a slightly different question than the one raised by Currie, it is just as valid of a question and is easier to apply. The laboratory only needs to calculate and report the TPU for the sample, something that most of them are already doing. Furthermore, DLs calculated in this way are more sample specific because they do not assume that the sample has the same detector background and, therefore, the same counting uncertainty as the blank. However, it does require that the TPU be appropriately calculated.

DLs calculated in this way have been shown to compare favorably with the DLs calculated by evaluating the probability that the result is not part of the blank population as long as the TPU is used in the derivation and not the counting uncertainty only. DLs calculated in this way are also easier to determine and implement by both the laboratory and the projects since only the TPU has to be reported. DLs calculated in either of these ways have resulted in no more than 5% false positive decisions. This shows that the effect of the Poisson distribution on the decision rate appears insignificant even at low background counting rates as seen in alpha spectrometry. This is likely due to the fact that the distribution becomes more Gaussian after background subtraction. The high false positive rates experienced by others are likely due to not incorporating all of the measurement uncertainty in the calculation of DL.

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# An Improved Method for the Determination of $^{235}$U and $^{238}$U in Urine By Inductively Coupled Plasma-Mass Spectrometry

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## Abstract

An improved method has been developed for the determination of $^{235}$U and $^{238}$U in urine by inductively coupled plasma-mass spectrometry. Matrix effects, spectral interferences, variations in nebulizer efficiency, etc., can significantly affect the results. The purpose of this study was to assess the feasibility of using an internal standard to account for the variations introduced by the above-mentioned effects. Various applicable internal standards were investigated. The acidified urine samples were diluted ten-fold prior to analysis. Natural and artificial urine samples were analyzed with this method. Consistent and accurate results were obtained when utilizing $^{236}$U as the internal standard. This paper will present the improved method and the results of these investigations.

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# Application of extended dynamic range Sector Field ICP-MS to the analysis of high uranium matrices

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When faced with the problem of the complete elemental analysis of a sample with high matrix concentrations, the analyst has traditionally been forced to apply multiple instrumental techniques depending on the concentration of the target analytes:

- Majors: XRF, AAS, ICP-AES
- Minors: ICP-AES, Q-ICP-MS
- Traces: Q-ICP-MS, SF-ICP-MS
- Ultra-traces : SF-ICP-MS

This leads to increased sample handling, complexity, time and cost. No single technique has been able to provide a complete elemental analysis.

Sector-field ICP-MS, with its intrinsic abilities to guarantee high elemental sensitivity and high selectivity, has become the technique of choice in many nuclear laboratories for the analysis of trace and ultra-traces concentrations in complicated sample matrices. With its characteristic flat-top peaks, it has also been used for high precision isotope ratio analyses of (for example) low concentrations of U in biomedical / environmental applications (e.g. DU in urine etc).

With the introduction of the ELEMENT XR that, unique in ICP-MS, incorporates a single Faraday detector in addition to the standard ELEMENT2 dual-mode (analog/counting) detector, the linear dynamic range available to the analyst with this instrument has been increased by 3 orders of magnitude to over $10^{12}$. With the high sensitivity and low background characteristics of SF-ICP-MS, this detection range is equivalent to <fg/g to > 1000 µg/g in solution mode.

In addition to the improvement in the measurable concentration in elemental analyses, the incorporation of the Faraday detector into the highest precision single collector ICP-MS enables the measurement of large isotope ratios (e.g. $^{234}U/^{238}U$) at sub ppm total U concentrations.

This presentation will summarize the implementation of this unique detector system in the ELEMENT XR and include examples of its in elemental and isotopic applications relevant to nuclear materials.

# New and fast analytical protocol for the determination of actinides in air filter samples by HPLC-ICP-MS

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**Abstract** – In case of a radiological / nuclear (R/N) emergency, the sample throughput of an analytical method would be a significant factor for rapid delivery of quantitative results that provide the level of contamination in various samples obtained from the emergency site. It is well known that for the determination of radionuclides with long half-life ($t_{1/2} > 2\ 000$y), mass spectrometry is a more rapid and convenient approach. This is also true for most of the actinides, which are long-lived and α-emitters, and usually require time-consuming sample preparation.

We have developed a chromatographic separation protocol for the measurements of actinides (i.e. Th, U, Np, Pu, Am) collected onto air filters. The chromatographic protocols are based on three steps: radioanalyte pre-concentration, matrix-constituents removal, and selective elution of the desired analyte. In order to increase the efficiency and reproductibility of the developed separation protocol, a high performance liquid chromatography (HPLC) was coupled with a sector-field inductively coupled plasma mass spectrometer (ICP-MS) through a high sensitivity sample introduction unit (APEX-Q). This presentation will describe the separation protocol, the automation procedure, and the figures of merits of the system for the analysis of Th, U, Np, Pu, and Am in spiked air filter samples. This protocol is part of the governmental initiative to improve Canada preparedness in case of a R/N emergency by providing rapid, accurate, and precise measurements.

UCRL-ABS-213685

# Age-related trend in elevated plutonium-239 measured by AMS in urine samples collected in 1998-2003 from Enewetak residents and Rongelap resettlement workers

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Under an IRB-approved human subjects protocol, a total of 377 urine samples were collected from 167 (male + female) residents of Enewetak Atoll during 2001-2003, and from 115 (male) Rongelap Atoll resettlement workers, together with field blanks prepared by identical methods substituting double-distilled $H_2O$ for collected urine. Extensive precautions were taken to avoid sample contamination, including physical isolation in an indoor facility during a uniform 24-h sample collection period. Measures of $^{239}$Pu subsequently made at the LLNL Center for Accelerator Mass Spectrometry (AMS) included estimates of combined measurement error (ME) due to chemical recovery and AMS isotope sampling. Measures from each multiply sampled individual were pooled, and participants on each atoll were grouped by age at sampling time into three ranges (<35, 35-44, ≥45 y, rounded down to nearest y), corresponding to low, medium and high levels of relative historical exposure to fallout due to atmospheric nuclear testing estimated using published UNSCEAR data. The $^{239}$Pu measures obtained from older age groups were significantly elevated compared to pooled blanks, as assessed by 2-tail Monte Carlo compound Mann-Whitney (CMW) tests that account for all ME (Figure 1). Across age groups (with blanks taken as age 0), the group-specific fractions of measures >0.35 μBq were found to be positively associated with age, ignoring ME ($p < 0.001$, by atoll-specific chi-square tests for linear trend). Accounting for all ME, the positive age-related trend for Enewetak is statistically significant (Figure 2). Observed trends in relation to age were stronger that those in relation to relative exposure to cumulative fallout. [This work was performed under auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory (LLNL) under Contract W-7405-Eng-48, with support from the LLNL Marshall Islands Program.]

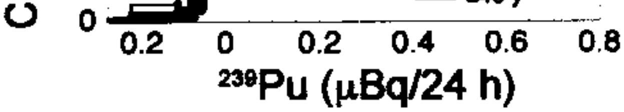

*Figure 1. Empirical probability mass functions (cmfs) for $^{239}$Pu in pooled field blanks vs. urine samples from all 285 study participants (each cmf is weighted by corresponding inverse-square ME sets). $^{239}$Pu is significantly elevated among all participants ≥35 y ($p < 0.035$, by CMW tests).*

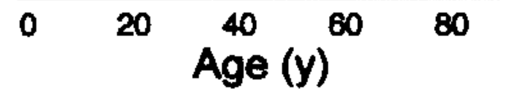

*Figure 2. Urinary $^{239}$Pu is significantly positively associated with age for all Enewetak ($n = 170$) participants, as assessed by 0-intercept weighted linear regression of all data pooled by age group, vs. corresponding average age, using $ME^{-2}$ as weights (solid horizontal line = 0 μBq/24-h, short-dashed lines = 95% confidence limits on slope). Error bars denote ±1 SEM; 95% confidence limits on the fitted intercept include 0.*

# What a Coincidence

## Developing a Rapid LSC Screening Method for $^{90}$Sr Contamination in Urine

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Strontium-90 is one of the most insidious fallout products of nuclear fission. Radioactive but difficult to detect due to its characteristics as a pure beta emitter, it is relatively long-lived with a ~29-year half-life. Its daughter yttrium-90 is another pure beta emitter, further complicating the detection of $^{90}$Sr. Strontium is chemically analogous Ca; thus, in the body $^{90}$Sr is deposited in bones and teeth, its radioactive isotopes causing long-term complications such as leukemia in humans. Current $^{90}$Sr bioassay methods require a few days to several weeks per sample and involve time- and labor-intensive chemical separations. This time frame is unacceptable for mass human screening in the event of the release and environmental dispersion of considerable quantities of $^{90}$Sr.

A rapid (~1-2 hours) method has been developed to screen human urine samples for $^{90}$Sr contamination via parallel Cerenkov and cocktail liquid scintillation counting. Determination is complicated by chemical and color quenching varying with different urine samples. Corrective quench curves can be quickly and simply generated on a sample-by-sample basis by splitting the sample and spiking one fraction with a radioactive isotope for comparison against an unquenched sample, reducing or eliminating the necessity of chemical separation processes. Due to its relatively low maximum beta energy, only ~0.5% of $^{90}$Sr activity will be counted via Cerenkov LSC, while >90% is counted via cocktail LSC. A calibration constant is calculated to allow for the comparison of these two measurements and thus $^{90}$Sr fraction in the urine is resolved. The method also allows for the resolution of $^{89}$Sr and $^{90}$Y from the energy spectra. This method is robust, allowing for rapid mass screening of the population affected by the dispersion of radiostrontium.

# ON-SITE COLLECTION, CONCENTRATION AND SEPARATION OF NUCLIDE SPECIFIC ANALYTES IN URINE

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A procedure and device are described that allow the on-site concentration and separation of nuclide-specific analytes in a one-time voiding of urine. Present collection methods require that bulk (one to two Liter) urine samples be collected on-site and shipped to a remote laboratory before the analytical procedure is begun. The transit time between shipping and the initiation of analysis allow many complex and irreversible reactions to transpire. In order to destroy the resulting matrix, the sample is treated aggressively with harsh chemicals in a time-consuming procedure to force the separation of the target nuclide. The paper describes a method, including the reaction vessel, into which the sample is introduced followed by the addition of acid and denaturing agents. Nuclide-specific resins added to the container separate and concentrate the analyte of interest by mass action. The resin is retained as the remaining sample is discarded. Sample volumes are reduced to approximately 20-30 mL. Upon receipt of the sample at the analytical laboratory, extraction of the target nuclide is straightforward and relatively rapid. Results of retention for various target nuclides as a function of acid and denaturing agent concentration are provided. Where feasible, mass spectrometers provide rapid sample turn-around as well as the quantification of extremely low concentrations of the target nuclide. Application to the emergency management of an uncontrolled release of radioactivity is discussed. Extension of the method and device coupled with on-site instrumentation is considered with estimated detection limits for the protection of first responder personnel and the affected population. Variations of the sample-collection device design and analytical method are protected by the U.S. Patent Office.

OBU-OLS-2005-0031

# Validation Process for Bioassay MDCs

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Since 2001, the WSRC Radiobioassay Laboratory has analyzed thousands of urine blanks and over 200 double blind spiked samples. This data has been used to validate the minimum detectable concentration (MDC) calculation in use by the laboratory. Examples of how the data has been used will be discussed for Pu-238, Pu-239/240, and Np-237.

UCRL-ABS-213684

# AMS Analysis of $^{239}$Pu in Archived Occupational Samples

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Under an IRB-approved human subjects protocol, accelerator mass spectrometry (AMS) was used to recover previously inaccessible information concerning short- and long-term excretion patterns of urinary Pu-239 for LLNL Pu workers who uniquely were both monitored periodically at LLNL via urine sampling over a period of decades and also had corresponding records archived at LLNL, in some cases for ≥20 years. Because the AMS methods used were substantially (50- to 300-fold) more sensitive than alpha spectrometry, excretion biokinetics were recovered from the archived LLNL alpha spectrometry discs to obtain measures pertaining to the biokinetics of human Pu-239 excretion with unprecedented sensitivity. AMS analysis was performed on residue from alpha spectrometry discs used historically to monitor Pu-239 excretion from a total of 11 current or former LLNL employees: 7 suspected of having ≥1 previous low-level occupational Pu-239 intake, and 4 thought to have had no such intake. AMS data obtained clearly confirmed occupational intakes for all workers suspected of previous exposure. AMS data for one worker time series provide evidence of an historical Pu-injection wound that most likely occurred 15-20 y prior to its recent discovery by body counting methods (Figure 1). AMS data obtained for this and other workers indicate that even extremely low-level occupational exposures to Pu-239 typically yield body burdens that remain readily detectable in urine analyzed by AMS, effectively marking any such worker for life—an observation that may have important occupational health and national/homeland security applications. [This work was performed under auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory (LLNL) under Contract W-7405-Eng-48.]

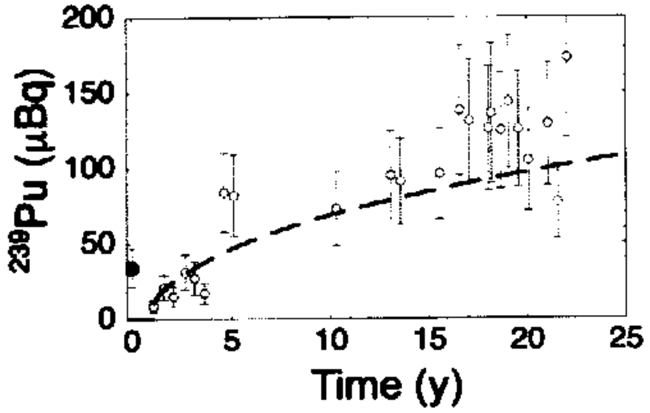

***Figure 1.*** Urinary $^{239}$Pu (points ±1 SE) detected by retrospective AMS analysis of archived sample plates prepared from urine samples obtained from a male worker by retrospective AMS analysis of archived bioassay plates, vs. relative time $t$, shown fit to three different SE$^{-2}$-weighted regression models. The linear fit shown (solid line) is 4.3 (±3.1) µBq + $t$ 5.96 (±0.583) µBq y$^{-1}$ ($\chi^2 = 18.7$, df = 21, p = 0.36; this fit excludes outlying solid point, $F_{1,21} = 5.68$, p = 0.027). A fit of the biokinetic model $A(t) = a\ U_{\mathrm{cIV}}(t-t_1)$, where $U_{\mathrm{cIV}}(t) = \int_0^t U_{\mathrm{IV}}(s)\mathrm{d}t$ and $U_{\mathrm{IV}}$ is the current ICRP biokinetic model for IV injection of Pu, is shown (bold long dashed curve) with estimates $t_1 = 1.06$ (±0.12) y and $a = 489$ (±37.4) ($\chi^2 = 32.8$, df = 22, p = 0.065). The fit to a similar model assuming Pu injection at times $t_0 = -10/365$ y and $t_1 = 4$ y is also shown (short dashed curve; $\chi^2 = 14.4$, df = 21, p = 0.85). All three models are statistically consistent with the AMS data.

# The HML's new whole body counter: an extension of the calibration using Monte Carlo simulations.

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## INTRODUCTION

The Human Monitoring Laboratory (HML), which operates the Canadian National Calibration Reference Centre for *In Vivo* Monitoring (Kramer and Limson Zamora 1994), has replaced its whole body counting system. The new whole body counting system is based on germanium detectors and its calibration and comparison with the previous counting system has been described elsewhere (Kramer and Hauck 2005).

This paper describes the extension of the calibrations using Monte Carlo simulations to provide information about the efficiency curves in energy regions for which no standards emit photons. The simulations also avoid the purchase of additional radionuclide standards and their subsequent disposal. Length restriction of this extended abstract precludes the inclusion of data tables and figure to illustrate the pints in the text

## METHOD AND MATERIALS

**The Detectors:** The new counting system consists of four 150% hyperpure germanium detectors mounted on a single Dewar of 68 L capacity whose rated holding time is 5 days. Each detector is approximately 94 mm in diameter and 105 mm in length. The P-type crystals have a dead layer of 700 μm and are encased in a 2.5 mm magnesium shield. Only the detectors, part of the counting bed, and the phantoms were included in the Monte Carlo simulations.

**Monte Carlo Simulations:** Photon energies were simulated over a wide range (60 to 2000 keV) with increments scaled according to the curvature of the efficiency curve. For example, in the region of the curve's knee the increment was 10 keV, whereas at the tail's end it was 250 keV or more. Two benchmark energies were chosen that correspond to radionuclide standards: 60 keV ($^{241}$Am) and 1836 kev ($^{88}$Y).

The detectors were modelled from data provided by the manufacturer and a 700 μm dead-layer was used for the germanium crystals. The BOMAB phantoms were modelled as described previously (Kramer et al 2002). The Lexan bed-support and the foam mattress were also included in the model. The MCNP descriptions of the BOMAB phantoms will be made available to interested parties and can be obtained by contacting the authors.

Each photon energy was run independently and was mono-energetic for each of the ten counting positions. Each run was set for $10^7$ histories (particles). The photons that interacted with the active germanium (not including the dead layer) in the detectors were tallied individually (bin width 1 keV) so that an individual detector efficiency was obtained for each configuration. A scan was obtained by averaging the ten counting positions and a four-detector array was simulated by simply adding up the individual detector tallies for a given configuration.

The simulations were run on four AMD64 3500+ computers (2.1 Gb Hz, 2Gb RAM). The total number of simulations run was 1860 and each run took approximately 24 minutes (total run time approximately 744 hrs).

The Monte Carlo code used for the simulations has been described in detail elsewhere (Kramer et al 2002). The authors of MCNP consider that a relative error on a predicted value calculated by the code of 0.1 - 0.2 suggests that the tally result is questionable (X5 Monte Carlo Team 2003). Tally results for which the relative error is above 0.2 are not likely to be meaningful; however, results are generally reliable for a relative error less than 0.1. The number of photons used in the simulations was $10^7$ so that the relative error varied from 0.007 to 0.094. Initially 40 keV photons had been included in the modelling, but the relative errors were all in excess of 0.2 so further data handling was discontinued.

**Phantom Size:** Measuring a subject, or a phantom, one has to consider two size variables that will affect the counting efficiency: height and weight. Previous work (Smith et al 1979) has found that the counting efficiency is proportional to $(\text{weight/height})^{1/2}$ in a whole body counting system.

## RESULTS AND DISCUSSION

**Calibration:** Contrary to the previous calibration methodology of the HML's whole body counter, the order of counting efficiency is no longer P4 > P10 > PM5 > PF > PM > PM95 but is now P10 > PM5 > PF > P4 > PM > PM95. The counting efficiency of the P4 phantom is slightly greater than the PF phantom at low photon energies, but at higher energies (>400 keV) the efficiency of the P4 phantom drops below that of the PF phantom and at still higher energies (>1700 keV) drops slightly below that of the PM phantom.

The reason for this lies in the scanning length. Previously, the HML had a variable scan length for its whole body counter so that scanning would commence at the subject's head and finish at the feet. The new whole body counter is not configured for variable scan length and each measurement covers the entire length of the counting chamber. The scans for the shorter phantoms, therefore, include differing lengths of "empty"bed. The largest section of "empty" bed being for the shortest phantom.

When the counting efficiency is optimised for scan length, the counting efficiency values, increase by a factor of 1.5 (1.52 to 1.54) for the P4 phantom at all energies, 1.17 for the P10 phantom, 1.06 for the PM5 and PF phantoms, and remain unchanged for the PM and PM95 phantoms.

**Minimum Detectable Activity (MDA) and Dosimetric Implications:** MDA values have been previously calculated for the Reference Male BOMAB phantom (Kramer and Hauck 2005). In the existing configuration (non-optimised scan length) of the HML's whole body counter, the P4 phantom has counting efficiencies slightly higher than that of the PM phantom. Assuming the background to be the same, because the counting time is the same for the two sizes, then the MDA's for children will be only slightly lower than that of a male corresponding in stature to that of the PM phantom. If the scan length were optimised (but the counting time kept the same), then the MDA values would be expected to drop by a factor of 1.5 for children corresponding in stature to the P4 phantom.

Previously (Kramer and Hauck 2005), the dosimetric implications of the MDA's were evaluated. An improvement of a factor of 1.5 in the MDA does little to improve the absolute sensitivity of the HML's whole body counter. $^{57}$Co, $^{137}$Cs, and $^{60}$Co can already be detected far below the dose limit (i.e., 20 mSv). An improvement of a factor of 1.5 for $^{241}$Am is still insufficient to make much of a difference, if it is detected then the committed effective dose will be between ~100 to ~150 mSv.

**Phantom Size:** The relationship of counting efficiency to size was investigatedfor the existing configuration (constant scan length) and for the optimised configuration (variable scan length). An arbitrary function that fits the data well for the existing configuration is given below:

$$Eff = a + \frac{b}{y} + \frac{c}{y^2} + \frac{d}{y^3}$$

where: $Eff$ = counting efficiency (cnt/photon), $y$ = the phantom's (or person's) size (weight in kg divided by height in metres)$^{1/2}$, and $a$ - $d$ are regression coefficients.

The data for the variable scan length of the HML's previous whole body counter was best fitted by the following expression:

$$Eff = e^{(a + b \cdot y)}$$

where: $Eff$ = counting efficiency (cnt/photon), $y$ = the phantom's (or person's) size (weight in kg divided by height in metres)$^{1/2}$, and $a$ and $d$ are regression coefficients.

This equation can be combined with the equation that described the efficiency as a function of energy to give the combined function that related counting efficiency, size, and photon energy:

$$Eff = e^{\left[a + by + \frac{(c+dy)}{Ln(x)} + \frac{(e+fy)}{\{Ln(x)\}^2} + \frac{(g+hy)}{\{Ln(x)\}^3} + \frac{(i+jy)}{\{Ln(x)\}^4}\right]}$$

where: $Eff$ = counting efficiency (cnt/photon), $x$ = photon energy (keV), $y$ = the phantom's (or person's) size (weight in kg divided by height in metres)$^{1/2}$, and $a$ - $i$ are regression coefficients.

The function gave predicted efficiencies that were in the range of 10% to -8% of the true value for the HML's previous whole body counter. Unfortunately, the decision to use a constant scan length, which is now based on operational simplicity and hardware restrictions, means that Eqn 3 cannot be applied to the current data.

The data corresponding to the constant scan length cannot be fitted well using Eqn 2. Using Eqn 1, the fit is good with predicted efficiency values being within 1% of the true value.

Combining Eqn 1 with an equation that describes the efficiency as a function of photon energy one obtains:

$$Eff = \left(a + \frac{b}{y} + \frac{c}{y^2} + \frac{d}{y^3}\right) \cdot \left(e^{\left(\frac{e}{x} + \frac{f}{x^2}\right)} - e^{\left(\frac{g}{x} + \frac{h}{x^2} + \frac{i}{x^3}\right)}\right)$$

where: $Eff$ = counting efficiency (cnt/photon), $x$ = photon energy (keV), $y$ = the phantom's (or person's) size (weight in kg divided by height in metres)$^{1/2}$, and $a$ - $i$ are regression coefficients.

An alternate version that works almost as well is given by:

$$Eff = \left(a + \frac{b}{y} + \frac{c}{y^2} + \frac{d}{y^3}\right) \cdot e^{\left(ex + f + \frac{g}{x} + \frac{h}{x^2} + \frac{i}{x^3} + \frac{j}{x^4}\right)}$$

where: $Eff$ = counting efficiency (cnt/photon), $x$ = photon energy (keV), $y$ = the phantom's (or person's) size (weight in kg divided by height in metres)$^{1/2}$, and $a$ - $j$ are regression coefficients.

And a third form is given by:

$$Eff = \left( a + \frac{b}{y} + \frac{c}{y^2} + \frac{d}{y^3} \right) \cdot e^{\left( e + \frac{f}{\ln\{x\}} + \frac{g}{(\ln\{x\})^2} + \frac{h}{(\ln\{x\})^3} + \frac{i}{(\ln\{x\})^4} \right)}$$

where: *Eff* = counting efficiency (cnt/photon), $x$ = photon energy (keV), $y$ = the phantom's (or person's) size (weight in kg divided by height in metres)$^{1/2}$, and $a$ - $i$ are regression coefficients.

The above equation predicts counting efficiency as a function of size and photon energy to within 10%, whereas the other two equations only predict to within 15%.

## CONCLUSIONS

The use of Monte Carlo simulations can fill in large gaps in an efficiency curve and help better define its shape.

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# Getting into the BOMAB Head

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In the event of a radioactive disaster, one of the biggest tasks is to estimate the radiation dosage received by people to determine the actions of emergency response teams. As part of these radiation dosage calculations, accurate measurements of the contaminated people require an estimated geometrical efficiency based on the measuring equipment of the human body. This implies that there is a need for a large number of human body standards (phantoms) to meet the quality criteria of the measurement. The purpose of this project was to find a reliable way to estimate the efficiency of gamma systems using the Monte Carlo computation, and to validate that efficiency by making measurements of a standard geometry. The final scope of the project is to create a standard human body phantom, to validate its theoretical efficiency based on a comparison of the Monte Carlo computation with the experimentally measured efficiency, and to calibrate existing phantoms.

A Bottle Manikin Absorption (BOMAB) phantom head spiked with Ga-67 was used for this project as a standard geometry. The radioactive BOMAB head is measured at a number of distances from HPGe detector, and the experimental efficiency for our gamma spectrometry system is determined. The same set of experiments is then modeled using the Monte Carlo N-Particle Transport Code (MCNP). The main task for the MCNP calculations is to create an accurate and realistic description of the measuring system (detector, source, and the environment). In this project, a method is proposed to define the object's geometry by using its CT scan data. A computer program called Scan2MCNP was used to convert CT scan data of the BOMAB head into a very accurate 3-dimensional computer model to be included into the MCNP input file (Figure 1). The project output will make it possible to measure similar geometries with unknown activity using the estimated efficiency by MCNP based on its CT images.

The theoretical and experimental results are compared. The results of the project that is currently in progress will be reported: 1) results between computations and measurements are consistent to better than 4 percent, 2) the uncertainties for both computations and measurements are better than 4 percent (k=1), 3) the major uncertainty for the MCNP computations was the description of the geometry of head-detector system, and 4) the major uncertainty for the measurements was the emission probability of the gamma-rays.

Figure 1- Computer Model of the BOMAB head as viewed in the Moritz program.

UCRL-ABS-212876

# The Heart Must be Removed Prior to In Vivo Lung Counting to Achieve Optimum Detection Efficiency of Low Energy Gammas

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The Lawrence Livermore National Laboratory Whole Body Counting program uses Canberra ACT II low energy germanium detectors for lung counting. Two sets of two detectors each, with one set positioned over the left upper torso region and the other set positioned over the right upper torso region. Each set of detectors is configured with a single liquid nitrogen dewar. LLNL has installed a multivariate positioning system to optimize positioning of each set of detectors, allowing for optimal positioning of the detectors on phantoms and human subjects.

Many physical aspects of the human body impact the efficiency of the counting system including the heart, rib cage, fat to muscle ratio thicknesses, and counting geometry. While many of these factors can be averaged across the entire chest region, the heart only impacts results from the left detector. This paper will focus on methods and results of an investigation to optimize the position and geometry of the left detector while minimizing the attenuation of low energy gammas caused by the presence of the heart. By performing measurements using multiple positioning of the left detector, LLNL has been able to establish a geometry where the heart has a minimized impact on lung counting efficiency.

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

# High Resolution HPGe Lung Counting
# Without Liquid Nitrogen

LAUR-03-5915

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## Abstract

For nearly 3 decades, high purity germanium (HPGe) semiconductors have been used as gammas spectrometers for the assay of radioactive materials in the lung. High purity germanium detectors are preferred because the semiconductor's properties yield gamma and X-ray spectra with peaks of minimal resolution, enabling positive identification of most radionuclides of interest, even in the presence of interfering radionuclides. The primary drawback associated with high purity germanium in a lung counting application is the use of liquid nitrogen, required to cool the detectors to temperatures which permit good charge collection (nominally 90 to 110 K). Some of the major concerns are the cost in time and materials to use and maintain a liquid nitrogen fill system, the inconvenience and hazards associated with frequent filling of detector dewars, and the perceived risks from potentially hazardous liquid nitrogen in close proximity to the subject. Over the years, numerous attempts have been made to employ mechanical coolers for HPGe detectors in lung counting applications, but all have proved too expensive, too noisy (resulting in unacceptable resolution degradation), or both. This paper describes the application of a new, low cost, low noise, low power commercially available cooler to lung counting HPGe detectors.

# The sliced BOMAB phantom - comparison with a homogeneous BOMAB phantom.

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## INTRODUCTION

The Human Monitoring Laboratory (HML), which operates the Canadian National Calibration Reference Centre for *In Vivo* Monitoring, previously developed the concept of a sliced **BO**ttle **Ma**nnikin **AB**sorber (BOMAB) phantoms which was tested using Monte Carlo simulations. The simulated phantom was made of solid high density polyethylene sliced into 2 cm sections in the transverse direction. Radioactivity was introduced using planar sources placed between the slices.

The response of the sliced phantom was compared to that of a conventionally designed phantom that had the activity dissolved in an aqueous solution and homogeneously distributed throughout the phantom. It was found that the two types of phantom performed almost identically, i.e., they had very similar counting efficiencies over a wide range of energies. It was concluded that a prototype phantom should be constructed and experiments carried out to validate the findings of the Monte Carlo predictions. This paper describes the construction of that sliced BOMAB phantom and compares its performance with a conventional BOMAB phantom on three whole body counting systems.

## METHODS AND MATERIALS

**The Phantom:** The phantom was constructed by Canus Plastics (Canus Plastics, 300 Lisgar Street, Ottawa, Ontario, K2P 0E2) from solid high density polyethylene sheets either nominally 1.90 cm or 1.27 cm thick and cut to shape to match the cross sections of the ten sections (head, neck, arms, chest, gut, thighs, and calves) of the HML's Reference Female phantom.

One major departure form the simulated phantom was the addition of connecting rods which are required to keep the slices of each of the BOMAB phantom's sections together. Each section has two rods to keep the slices and inserts in place. Each polyethylene rod is 1.27 cm in diameter, threaded at both and is the length of the section it holds together. To avoid the connecting rod protruding beyond the end of the section one of the end slices has a recessed groove were the nuts are attached to the rods. The other end of the rod screws directly into a thread slice. Figure 1 shows these features.

**The Planar Inserts:** Tracings were taken of one slice from each of the 10 sections of the sliced BOMAB phantom and a cardboard template was created. The templates were used for the production of the filter paper inserts that were spiked with activity. The filter paper inserts (Whatman No. 54 was used because of its wet strength and chemical resistivity) were labeled with a layer number and the section to which it belonged. Holes were cut into the filter paper in the areas where the connecting rods would pass through the planar source leaving a small gap so there would be no contact.

The inserts were then glued inside one half of a laminating pouch using a glue stick. The laminating pouches (Ibico clear laminating film pouch - Grand and Toy, Ottawa, Ontario), which are 21.6 cm x 28.0 cm, not could hold an entire piece for the chest and gut sections. These large sections were, therefore, manufactured in halves. For the smaller sections (i.e., arm and calves) several filter papers could be inserted into the pouch at once and adequate space was left between them so that the laminated inserts could be cut apart.

A standard solution ($^{57}$Co, $^{60}$Co, and $^{137}$Cs) was prepared from commercially (Amersham, Oakville, Ontario) purchased radionuclide solutions of known concentration. The desired total activity for each source was determined and an acidic solution (0.5 mol $L^{-1}$ HCl) containing this amount of activity was prepared in a 10 ml volumetric flask and transferred to a "baby bottle"which allows greater control of the drop size. The activity was deposited on the filter paper by wetting the filter paper with a series of small overlapping dots. The wetting commenced on the outer edge of the insert and worked in towards the middle until the paper was entirely wetted.

The weight of the solution added to the insert was determined by weighing the baby bottle before and after the addition. Once the entire filter paper was wetted and the desired activity deposited, the filter paper was allowed to air-dry in a fumehood. After drying, the pouch was closed and the filter papers were laminated (Ibico model PL-260 IC desktop pouch laminator - Grand and Toy, Ottawa, Ontario) and the planar inserts trimmed.

The inserts were loaded into the phantom as shown in Fig. 2 and the connecting rods were secured. The final activity loading in the sliced phantom was 9.5 kBq $^{57}$Co, 4.5 kBq 60Co, and 4.4 kBq $^{137}$Cs (23 Jun 2004).

**Comparison with a homogeneous BOMAB Phantom:** The HML replaced its whole body counter during the construction and characterization of the sliced BOMAB phantom so that it was possible to measure it on two different counting systems in the HML.

## RESULTS AND DISCUSSION

**The Phantom:** Table 1 summarises the physical parameters of the sliced phantom and also shows how many thin sheets were used to achieve the correct heights of each section. The total height of the phantom is 161.8 cm. Each laminated insert is 0.03 cm thick, so that the height increase due to the inserts varies from 0.1 cm (neck) to 0.8 cm (arms) and these increased heights are the values shown in Table 1. The total weight of the phantom is 50.4 kg. The target height and weight for Reference Female (ICRP 2002) are 163 cm and 60 kg. The heights agrees well; however, due to the lower density of polyethylene compared with bone and muscle the phantom is slightly light weighted.

**Comparison with a homogeneous BOMAB Phantom:** The bias obtained on the HML's old WBC, expressed as the difference of the sliced BOMAB's counting efficiencies from those of the homogeneous BOMAB phantom, was within ± 8% (for all energies measured) with the average values for each array configuration being ± 3%. Similarly, the bias obtained on the HML's new whole body counter was within ± 5% with the average across all energies being less than 3%.

**Design Modifications:** The Monte Carlo simulations tested a phantom that had no connecting rods. The presence of these rods in the manufactured phantom could perturb the performance characteristics of the sliced BOMAB phantom as there are now small inactive areas within the sections. As the rods are made of the same material as the slices no additional attenuation is expected. The experimental data showed that the sliced phantom agrees well with the homogeneous BOMAB indicating that the design modification was of little importance.

## CONCLUSION

The HML has designed and manufactured a new type of BOMAB phantom. The experiments in this work and the previously published work show that this phantom performs equivalently to a female BOMAB phantom whose activity is dissolved in water and homogeneously distributed throughout each section. The advantage of this phantom is that the sources are solid, sealed, and cannot leak activity. The new phantom is ideal for uses that involve shipment, such as for an intercomparison exercise.

**Table 1:** Dimensions of the sliced BOMAB phantom. 2a = semi-major diameter, 2b = semi - minor diameter, h = total height of section, # pieces is the total number of slices in the section, # thin pieces is the number of slices that are 1.27 cm thick (the others are 1.9 cm)

| Section | 2a (cm) | 2b (cm) | h (cm) | Weight (g) | # Pieces | # Thin pieces |
|---|---|---|---|---|---|---|
| Left Calf | 11.6 | 11.0 | 37.5 | 3610.5 | 20 | 2 |
| Right calf | 11.6 | 11.0 | 37.5 | 3610.0 | 20 | 2 |
| Left Thigh | 14.9 | 13.7 | 37.9 | 5811.5 | 20 | 2 |
| Right Thigh | 14.9 | 13.6 | 37.7 | 5796.0 | 20 | 2 |
| Gut | 33.3 | 18.2 | 19.0 | 8568.0 | 10 | 1 |
| Chest | 27.3 | 18.5 | 39.8 | 15462.5 | 21 | 1 |
| Neck | 13.4 | 12.5 | 9.0 | 1118.0 | 5 | 1 |
| Head | 17.4 | 13.4 | 18.8 | 3269.0 | 10 | 1 |
| Right Arm | 8.6 | 8.6 | 55.0 | 3135.0 | 29 | 2 |
| Left Arm | 8.7 | 8.6 | 55.2 | 3133.5 | 29 | 2 |

**Figure 1:** The sliced BOMAB phantom showing the connecting rods in place.

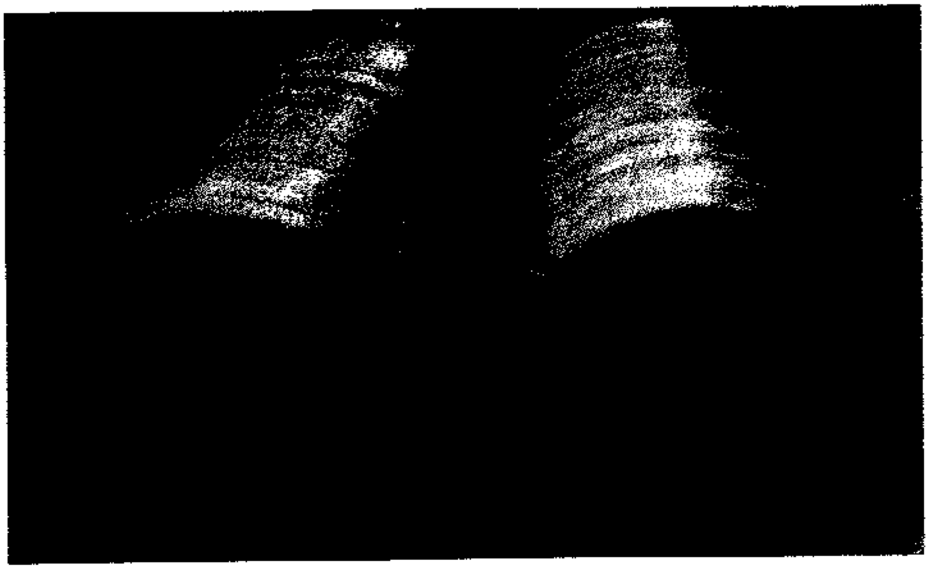

**Figure 2:** The sliced BOMAB phantom showing the planar sources being inserted into the head section.

# Development of Autonomous Unmanned Vehicles for Radiation Surveys and Sampling[1]

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Research is being conducted at the Idaho National Laboratory (INL) to develop unmanned autonomous radiological sampling and surveillance capabilities. The Environmental Services Project and Human, Robotics, and Intelligent Systems Department started collaborating in August 2000. Initial development was an unmanned aerial vehicle (UAV) to sample airborne radioactivity during range fires and abnormal events and perform aerial surveillance using on-board photographic and video equipment. Testing includes both fixed-wing and rotary airframes. Additional development is being performed on unmanned ground vehicles (UGV) and UAVs to perform remote radiological surveys using plastic scintillators and high-resolution gamma spectrometers.

The air robotic systems have already been used in multiple field deployments and operations. Recently, the remote aerial sampling and surveillance systems were used to take Wastewater Land Application Permit (WLAP) required aerial photographs of the application areas and high-resolution digital images of the INL Vadose Zone Research Park project for three dimensional mapping. Remote aerial sampling and survey equipment is now being developed and tested at INL using rotary wing vehicles with GPS navigation and small field-hardened portable detector systems to evaluate their usefulness for routine surveys and characterization of remediation sites.

Developing these capabilities can greatly reduce the cost and risk associated with these activities. Addition advantages include:

- Reduced manpower to perform the survey
- More accurate, complete, and reproducible results
- Reduced radiation exposure to personnel
- Ability to sample in hazardous and contaminated environments
- Reduced environmental impacts
- Reduced fire hazards associated with conventional survey vehicles
- Minimizing interference with remediation efforts.

As remote aerial sampling and surveillance technology becomes more accepted by end users, the technology baseline will move forward. As a result, the technology will need to be autonomous and easy to use. INL is developing the following additional capabilities:

- Integrating advanced technologies for detecting and avoiding obstacles, allowing safe operations during autonomous modes
- Improving user application software, allowing full intelligent autonomous operations
- Fully automating in-situ isotopic identification for increased efficiency and optimal data gathering

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[1] Work supported by the U.S. Department of Energy, Assistant Secretary for Environmental Management, under DOE-NE Idaho Operations Office Contract DE-AC07-05ID14516.

- Developing and integrating chemical contaminant detection capabilities
- Developing homeland security applications.

Developing these capabilities will advance the technology into the forefront of environmental surveillance and intelligent cooperative robotics. These state-of-the-art technologies will help DOE, Environmental Management, and other government and private agencies accomplish their mission to assess environmental impacts and protect the general public in a safe and timely manner.

# RADON MONITORING AT THE METROPOLITAN WATER RECLAMATION DISTRICT OF GREATER CHICAGO

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## ABSTRACT

Radon-222 is a colorless, odorless, and tasteless naturally occurring radioactive gas. It is produced by the radioactive decay of radium-226. The exposure of radon increases the risk of lung cancer at workplaces that have high radon levels. It is the second leading cause of lung cancer in humans.

The primary sources of radon exposure are soil, rock, and groundwater. There are certain geographical areas in the U.S. where relatively high concentrations of radium occur in groundwater. Many communities depend upon groundwater as their source of drinking water. Some of these water supplies have radium levels that exceed the drinking water standards. The community water systems remove radium from groundwater to comply with the drinking water standards, and discharge the residuals into the sewer system. The local water reclamation plants treat the raw sewage from these sewers to remove contaminants before discharging the effluent to surface water. The radium removed by the wastewater treatment process is concentrated in sludge. Radon may accumulate in wastewater treatment plant buildings at concentrations substantially greater than that prevailing outdoors. There may be potential for radioactive exposure of the treatment plant workers by inhalation of radon gas, resulting in a radiation dose to internal organs and tissues.

The Metropolitan Water Reclamation District of Greater Chicago (District) conducted a study to evaluate the exposure to radon of its employees. The radon concentration was measured at four water reclamation plants (WRPs) and at one tunnel and reservoir plan (TARP) location of the District. A long-term test was conducted using alpha track detectors for over 90 days. This study presents the results of radon monitoring at various locations within the District's four WRPs and one TARP location.

## INTRODUCTION

Radon-222 is a colorless, odorless, and tasteless naturally occurring radioactive gas. It is produced by the radioactive decay of radium-226, which is a decay product of uranium, and is found throughout the earth's crust. Radon has a half-life of 3.8 days, and it breaks down to a series of other radioactive elements. Unlike radon these elements are chemically active and electrically charged. These particles can adhere to air ducts in the lungs, emit ionizing radiation that damages the lung tissue, and produce cancer. Radon is the second leading cause of cancer in humans. It is estimated that 12% of lung cancer deaths in the U.S. are linked to radon (BEIR VI

1999). The exposure to radon increases the risk of lung cancer at workplaces that have high radon levels (Lubin et.al. 1994, 1995).

The primary sources of radon exposure are soil, rock, and groundwater. According to a U.S. Geological Survey/EAA Report (USGS 1998), "Radium is present in higher concentrations in some states, such as north-central states including southern Minnesota, Wisconsin, northern Illinois, Iowa, Missouri and southern states from Georgia to New Jersey". Elevated levels of radium-226 were found in certain groundwater systems of northern Illinois (Robert T. Key 1999). Many communities depend upon groundwater as their source of drinking water supplies. Some of these supplies have radium levels that exceed the drinking water standards for radium-226. These community water systems remove radium from groundwater to comply with the drinking water standards, and discharge the residuals into the sewer system. Naturally occurring radioactive material may also enter the sewer system with water infiltration or stormwater runoff into the collection system. Another general source of radionuclides in the environment is technologically enhanced naturally occurring radioactive material (TENORM). The TENORM may be introduced to the sewage system from groundwater and surface water, plants, foods, as well as from potential industrial discharges. Contaminants from raw sewage are removed at the WRPs by various wastewater treatments, such as physical, chemical, and biological processes. Reconcentration of contaminants can occur during sludge treatment (Ainsworth, et. al. 1994). Sludge processing techniques that may contribute to reconcentration include digestion, dewatering, and incineration.

The United States Environmental Protection Agency has set 4.0 pCi/L as the radon action level in air. The Illinois Emergency Management Agency, Division of Nuclear Safety (IEMADNS) has produced and published a brochure on "Radon in Cook County Area." In its study IEMADNS found that 17% of the homes in Cook County had indoor levels of 4.0 pCi/L of air or greater.

The District is located within the boundaries of Cook County, Illinois. It is one of the largest municipal treatment agencies in the U.S. and operates seven WRPs. All these WRPs are situated in the northern part of Illinois. It serves an area of 872 square miles including the City of Chicago and 125 suburban communities. The District treats 1500 million gallons per day (MGD) of wastewater from northern Illinois at its seven WRPs. The wastewater may transport radium either dissolved in the water or attached to suspended solids in the water. The District treats raw sewage containing radium to remove contaminants in order to ensure adequate water quality before the effluent is discharged to surface water. The radium removed by the wastewater treatment process is concentrated in sludge. Radon may accumulate in buildings during wastewater treatment at concentrations substantially greater than that prevailing outdoors. There may be a potential for radioactive exposure of the treatment plant workers by inhalation of radon and its progeny in both wet and dry conditions resulting in a radiation dose to the internal organs and tissues.

This study was conducted to measure radon concentration inside the District WRPs and to evaluate the exposure to the District employees. The degree of exposure depends on the configuration of the POTWs in terms of room size, ventilation, etc. Radon measurements were taken at locations where solids material accumulates including the grit chamber, and solids concentration buildings.

## MATERIALS AND METHODS

Alpha tract detectors, Radtrack, from Landauer, Inc. were used to measure radon concentration. The testing was conducted by an Illinois licensed radon measurement professional from Occusafe, Inc. Radon monitoring was conducted at the Stickney, Calumet, Lemont, and Kirie WRPs and at the Mainstream Pump Station. The devices were placed at various locations of the WRPs according to the IEMADNS testing protocol. Tests were conducted in frequently occupied rooms with substantial ground contact in the buildings and in areas where the potential presence of radon was possible. This included office areas, pump house, screening buildings, grit chambers, sludge holding tanks, and sludge loading buildings. Measurements were also made in several outdoor locations to measure radon emitted from various stages of wastewater treatment processes, such as aeration tanks and sludge digesters. The detectors were placed in these areas in a way that limited exposure to the sun and precipitation. A control device was placed outside in a grassy area to compare radon concentration levels that were not impacted by the operations.

The detectors were placed for a period of over 90 days. At the end of the collection time the detectors were sent to Landauer, Inc. for radon measurement. Duplicate radon measurements were carried out for at least 10% of the samples. Field blanks were deployed for at least 5% of the total number of devices to measure background levels.

## RESULTS AND DISCUSSION

The radiological monitoring data of the District's WRPs indicates elevated levels of radium-226 in Lemont WRP sewage sludge. The yearly average concentration of radium-226 in Lemont WRP sewage ranged from 44.9 pCi/g dry weight (dw) to 86.8 pCi/g dw from 1997 to 2004, whereas the Stickney, Calumet, and Hanover Park WRPs, it ranged from 2.1 to 5.5 pCi/g dw during the same period. The Lemont WRP sludge is transported by trucks to the Stickney WRP for further treatment. It is mixed with Stickney WRP sludge, treated, and sent to the solids drying sites before disposal. The dilution process considerably decreases the concentration of radium-226 in the final biosolids product. The radium-226 concentration found in the District's biosolids do not pose any significant risk to human health or the environment. Radon-222, a decay product of radium-226 may accumulate in buildings in concentrations substantially greater than those prevailing outdoors. A long-term test was conducted at the District facilities for the radon measurement because it provides a reading that is more likely to yield the year-round average radon levels. Radon samples were collected from various locations of four WRPs and one TARP.

### Stickney WRP

Radon testing was performed at the Stickney WRP from October 21, 2003 through January 27, 2004. A total of 18 alpha track detectors were placed including 13 single devices, 2 duplicates and 1 blank for 98 days. Some of the locations tested were in excess of 30 feet below ground.

Radon levels were well below the EPA recommended action level of 4.0 pCi/L in all locations tested and ranged from 0.3 to 0.7 pCi/L. The highest concentration of radon (0.7 pCi/L) was found at the grit chamber. Eleven of the sampling locations had radon concentrations of 0.3 pCi/L.

### Calumet WRP

Radon testing was performed at the Calumet WRP from June 26, 2003 through October 7, 2003. A total of 28 alpha track detectors were placed including 23 single devices, 2 duplicates, and a blank for 102 days.

Radon levels were well below the EPA recommended action level of 4.0 pCi/L in all locations tested and ranged from 0.3 to 2.6 pCi/L. The highest concentration of radon (2.6 pCi/L) was found at the coarse screen area. Fifteen of the sampling locations had radon levels of 0.3 pCi/L.

### Lemont WRP

Radon testing was performed at the Lemont WRP from May 28, 2003 through August 28, 2003. A total of 19 alpha track detectors were placed including 14 singles, 2 duplicates, and one blank for 92 days.

Radon levels were below the EPA recommended action level of 4.0 pCi/L in all of the locations tested except for two. The two areas that had radon levels above action level were the lowest level of the old pump house, 4.3 pCi/L, and the lower sump pump room in the main building, 10.8 pCi/L. The workers do not routinely occupy these two areas. They will have to work well over 8 hours per day every day for a year to exceed the action level. The average exposure of these workers is only approximately 5 hours per week and adequate ventilation is provided during this time.

### Kirie WRP

Radon testing was performed at the Kirie WRP from October 23, 2003 through January 29, 2004. A total of 15 alpha track detectors were placed including 12 single devices, one duplicate, and one blank for 98 days.

Radon levels were well below the EPA action level of 4.0 pCi/L in all the locations tested and ranged from 0.3 to 2.8 pCi/L. The highest concentration of radon (2.8 pCi/L) was found at the airlift station.

### Mainstream Pump Station

Radon testing was performed at the Mainstream Pump Station from May 28, 2003 through August 28, 2003. A total of 11 alpha track detectors were placed including 6 single devices, 2 duplicates, and one blank for 91 days.

Radon levels were below the EPA recommended action level of 4.0 pCi/L in all the locations tested and ranged from 0.3 to 0.8 pCi/L. The highest concentration of radon (0.8 pCi/L) was found at North Pump House. Seven sampling locations had radon level of 0.3 pCi/L.

## CONCLUSION

The results of this study show that the concentration of radon at all the tested locations of the District WRPs were less that the U.S. EPA recommended action level of 4.0 pCi/L annual average exposure except for two locations. The radon level at the lower sump pump room at the main building of the Lemont WRP was 10.8 pCi/L and at the old pump house was 4.3 pCi/L. The workers do not normally occupy these two areas. The data show that the radon levels in the air of the WRPs do not present elevated exposure to the workers.

It is concluded from this study that the radon concentration at the WRPs is not likely to have any adverse effect on the health of the District's workers.

## ACKNOWLEDGMENTS

The authors wish to thank the staff of General Administration for their assistance in conducting radon monitoring at the District.

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# Rapid Emergency Method for Actinides and Sr-89/90 in Water Samples

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There is also a growing need to have available rapid methods to assess actinides and strontium –89/90 levels in environmental samples for emergency preparedness reasons (1, 2). A new, faster actinide and strontium 89/90 separation method has been developed and implemented to improve the productivity, reduce labor costs and add capacity to this laboratory (3). The method can be used as a rapid method for emergency preparedness or for routine analyses. This method uses stacked TEVA Resin ®, TRU Resin® and Sr-Resin® cartridges from Eichrom Technologies (Darien, IL, USA) that allows the rapid separation of plutonium (Pu), neptunium (Np), uranium (U), americium (Am), curium (Cm) and thorium (Th) using a single multi-stage column combined with alpha spectrometry. By using vacuum box cartridge technology with rapid flow rates, sample preparation time is minimized. Thorium and curium are often analyzed separately due to the interference of the daughter of Th-229 tracer, actinium (Ac)-225, on curium isotopes when measured by alpha spectrometry. This new method also adds a separation step using DGA Resin ®, (Diglycolamide Resin, Eichrom Technologies) to remove Ac-225 and allow the separation and analysis of thorium isotopes and curium isotopes at the same time.

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3. Maxwell, S. L., "Rapid Column Extraction Method for Actinides and Sr-89/90 in Water Samples", *Journal of Radioanalytical and Nuclear Chemistry*, in press

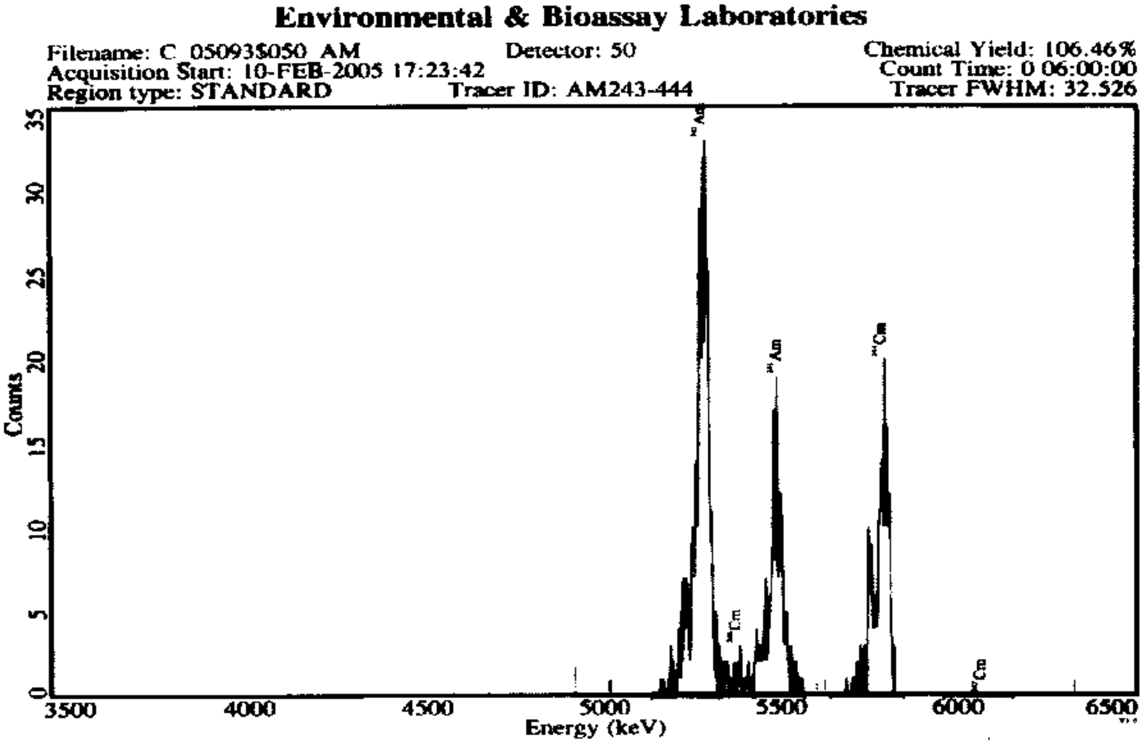

Figure 1 Alpha Spectra showing Am and Cm isotopes with Th-229 daughter (Ac-225) removed

# Investigation of radioactivity in waters in Luxemburg by follow-up radiochemical method

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In order to identify main related parameters concerning isotope concentrations and geological formations and to monitor the natural levels of naturally occurring radionuclides in ground waters, a survey of radionuclide content, chemical components, and some physical parameters was performed in 2002-2004 on the territory of Luxembourg.

More than 150 different samples were analysed for radium, radon and uranium, around 50% of them for tritium and only 30 for lead and thorium content. In fig. 1 are shown the sampling locations.

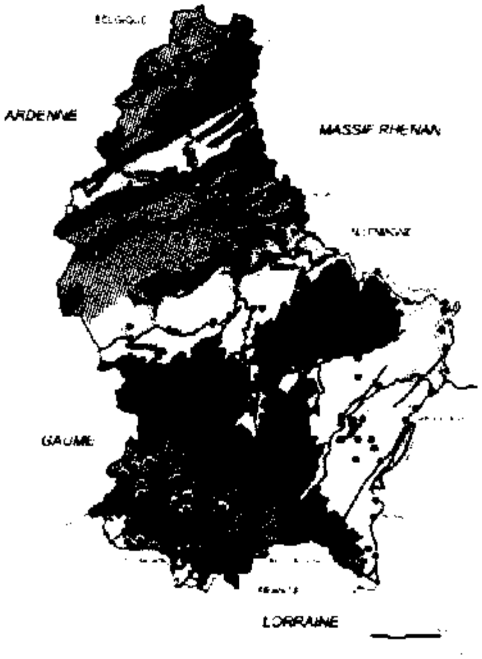

Fig. 1: Water sampling points

The surface water was collected mainly from the northern part of the country. The underground samples, more than 116, represent tapped sources and drilled well sources for potable drinking water.

Investigations were done to optimise the analysis conditions for natural radioisotope determination of the different underground and surface waters.

A time-saving and sensitive procedure of step by step analysis of the different radio-isotopes (follow-up method) for monitoring low concentration (activities) for $^{222}$Rn, $^{3}$H, $^{226}$Ra and $^{238}$U, $^{234}$U, $^{235}$U, as well as thorium isotopes and $^{210}$Pb/$^{210}$Po in water samples has been devised. Through the combination of co-precipitation and extraction chromatography effective radiochemical separation of the components was applied. After estimation of radon and tritium the rest of the samples were spiked with chemical recovery tracers. Figure 2 shows the scheme of the applied procedure.

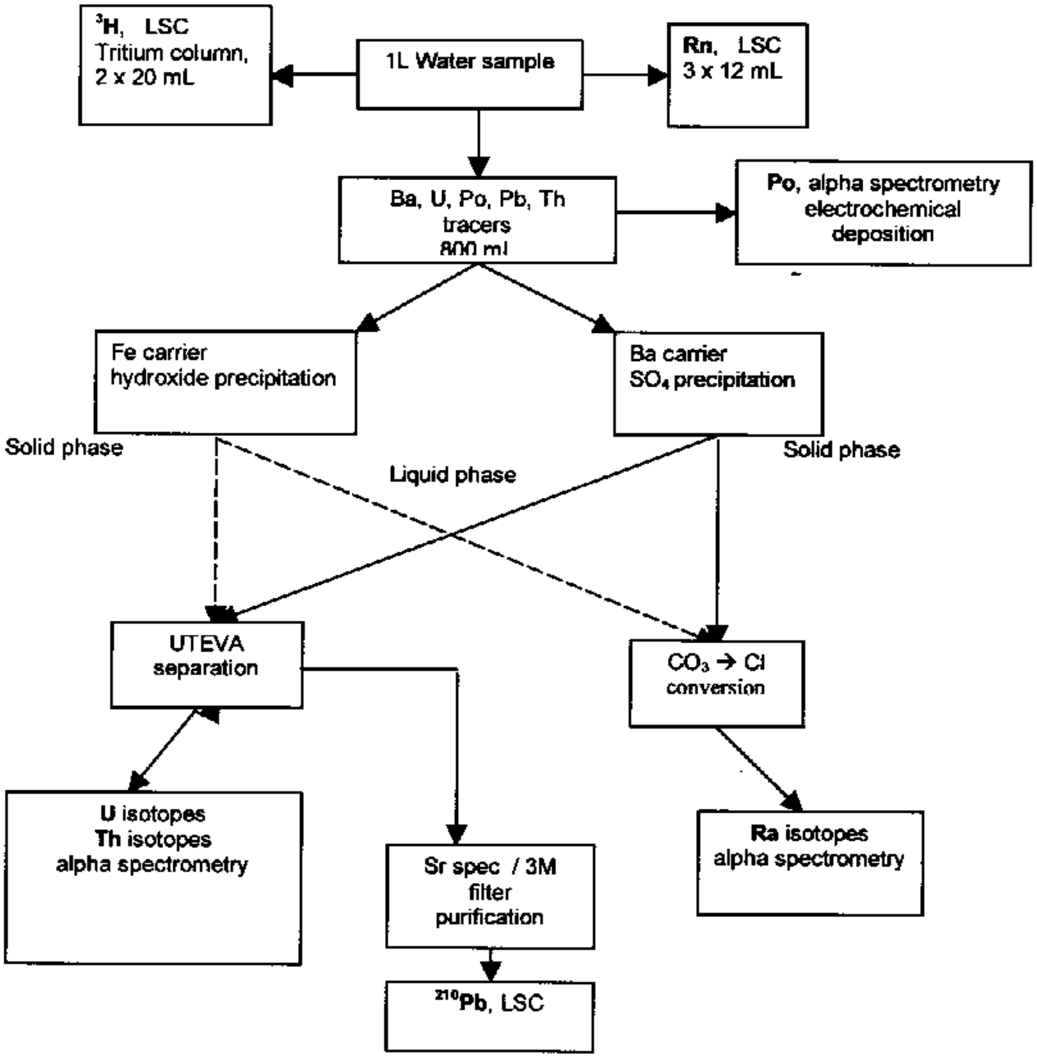

Fig. 2: Flow chart of chemical separation

Uranium, polonium and thorium activities were determined by alpha spectrometry and radium, tritium and lead by LSC, respectively.
The minimal detectable activities obtained were 0.83 mBq/L for $^{222}$Rn, 0.78 mBq/L for $^3$H, 0.6 mBq/L for uranium isotopes, 0.29 mBq/L for thorium isotopes, 2.5 mBq/L for $^{210}$Pb, 0.2 mBq/L for $^{210}$Po and 7 mBq/L for $^{226}$Ra.

A strong quality assurance procedure was applied. It includes double analysis of 10% of all the samples. Twenty of the samples were analysed by at least two methods. The results of the "follow-up" methods were compared with the others, obtained by different techniques. The comparisons were made for: a) radium alpha spectrometry on MnO$_2$

disks or filtered sulphate precipitation; b) uranium the liquid-liquid extraction procedure and LSC; c) tritium standard distillation procedure and c) radon similar procedure on portable LSC device. For radon all samples analysis were triple replicated.
The advantage of the chemical procedure is the relative low sample volume. One litre of water is sufficient to perform the analysis. The follow-up separation of different radionuclides is environment and health preserving as well time efficient. All procedures take only some hours and do not produce radioactive or organic wastes.
The obtained data were compared with the results from chemical analysis for major ions and geological features. Most samples were calcium-magnesium hydrogen carbonate type waters with high contents of iron, but significant correlations in between them could not be shown. This suggests independence among them. However the main influence on the radionuclide distribution is due to chloride ions.
Some explanation may be related to the surprising fact that almost all water contained iron and manganese in ratio 1:1.

Fig. 3: The map shows the distribution of radon and radium in sources and wells

The concentrations of radium vary from 1 to 140mBq/L, without taking into account the deep artesian wells Born and Rosport, where radium concentrations are as high as 200mBq/L.
In the different geological units (Triassic limestone, Triassic sandstone and the Palaeozoic basement) radium and radon concentrations are always anti-correlated. The waters deriving from the Palaeozoic part are slightly mineralised (elec. cond. 50 to 150 µS/cm) in comparison to the waters from the Mesozoic parts (elec. cond. 400 o 750 µS/cm). Nevertheless, radon concentrations are lower in the latter due to lower uranium-series elements in the rock matrix.
An analysis of the relation between radium and oxygen content shows higher radium concentrations associated with lower oxygen content. Waters high in oxygen have low

radium concentrations and vice versa. In oxygen rich waters, radium co-precipitates with ion and manganese to oxide and hydroxide minerals. This means that elevated radium concentrations are a signature of anoxic waters. In the case of waters from the northern part of Luxemburg (Palaeozoic formations) deep anoxic water are higher in radium content, which is precipitated when the environment changes to oxidizing conditions. This leads to hydroxide deposits in the upper parts of the aquifer where radium enrichment in the aquifer matrix is suggested. As a consequence the radium concentrations in water degrease in the same sense as the emanation product, radon, increases.

Fig. 4: Geological Map with the distribution of $^{234}$U and $^{235}$U.

In figure 4 the spatial distribution of uranium isotopes in the sources and wells is represented. The uranium isotopic concentrations are in the same order as the ones of radium. We obtained fairly low concentrations of uranium for the Liassic and Devonian geological units. A detailed study of uranium isotopes and their relations to thorium and lead concentration is in process.
Table 1 shows some results of this study.

Tab. 1: Results for uranium, thorium and lead in some water samples.

| Name | U-238 mBq/l | U-235 mBq/l | U-234 mBq/l | Pb-210 mBq/l | Th-232 mBq/l | Th-230 mBq/l | Th-228 mBq/l |
|---|---|---|---|---|---|---|---|
| Hersberg-40 | 12.31±0.07 | 2.45±0.02 | 13.71±0.11 | 29.5±0.08 | 5.18±0.03 | 3.37±0.06 | 7.59±0.09 |
| Hersberg-37 | 6.01±0.11 | 0.82±0.03 | 5.33±0.13 | 22.2±0.07 | 14.46±1.3 | 3.42±0.06 | 15.9±1.38 |
| Vianden | 1.62±0.03 | 0.14±0.02 | 0.38±0.02 | 0.47±0.03 | 7.72±0.09 | 3.51±0.05 | 6.78±0.08 |
| Haute-Sure | <0.285 | <0.165 | <0.215 | 22.5±0.07 | 5.91±0.08 | 3.86±0.07 | 8.08±0.10 |
| G235 | 1.10±0.04 | 1.27±0.04 | 7.74±0.31 | <0.03 | 3.04±0.03 | 4.94±0.03 | 2.88 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| G236 | 84.46±0.59 | 6.35±0.06 | 140.30±0.98 | <0.03 | 6.53±0.04 | 10.26±0.06 | 7.94 |
| G237 | 15.09±0.57 | 1.59±0.06 | 10.11±0.55 | <0.03 | 7.28±0.04 | 4.37±0.04 | 6.63 |
| G238 | 38.23±0.88 | 1.79±0.06 | 39.06±0.9 | <0.06 | 2.950.02 | 6.50±0.09 | 4.06 |
| G239 | 29.00±1.39 | 1.770.10 | 76.93±3.64 | <0.03 | 1.080.02 | 2.04±0.02 | <5.30 |
| G240 | 54.28±0.90 | 2.75±0.06 | 70.74±1.17 | 0.13±0.09 | 1.09±0.02 | 4.08±0.10 | <7.11 |

Only 40 samples were analysed for tritium content. They don't show values exceeding the natural background. Further correlation with dissolved oxygen and geology is to be done. The values vary in the range of 2 – 7 Bq /L.

# EFFECT OF QUENCH ON ALPHA/BETA PULSE SHAPE DISCRIMINATION OF LIQUID SCINTILLATION COCKTAILS

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## ABSTRACT

The effects of alpha and beta charged particle energy, various quenching agents, liquid scintillation cocktails, and liquid scintillation counters have on alpha/beta pulse shape discriminating liquid scintillation counting were quantified. Alpha emitting radionuclides, $^{239}$Pu and $^{241}$Am, and beta emitters, $^{90}$Sr and $^{99}$Tc, were investigated to quantify the effect of charged particle energy as well as nuclide dependence on alpha/beta pulse shape discrimination. The effects of chemical and color quenching agents, nitromethane, nitric acid, and yellow dye on alpha/beta pulse shape discrimination of Optiphase 'HiSafe' 2 and 3 (PerkinElmer™) and Ultima Gold™ AB liquid scintillation cocktails were determined. The prepared samples were counted on the Perkin-Elmer Wallac 1414 alpha/beta pulse shape discriminating liquid scintillation counter. It was found that for the same level of quench, as measured by the external quench parameter, the quench agents influenced the pulse shape discrimination parameters differently. The radiation energy as well as the radionuclide also affects alpha/beta pulse shape discrimination. By comparison with the Packard 3150 AB, the Wallac 1415 exhibited better pulse shape discrimination capability under the same experimental conditions.

# Magic or Science? ~$^{90}$Sr efficiency calibration data reduction

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$^{90}$Sr is one of the nuclides which have been determined routinely by many laboratories. It is pure beta emitter having short-life daughter, $^{90}$Y that is also a pure better emitter. This makes $^{90}$Sr analysis more difficult. When $^{90}$Sr is measured by proportional counter, one very important factor, which needs careful evaluation, is the detector calibration. Calibration is typically done by systematically changing the amount of Sr carriers to see the change of the efficiencies. Of course, there are many uncertainties associated with the efficiency, and these uncertainties should be propagated with the uncertainty of the analytical results.

In this study, we will discuss how we have calculated the uncertainty of efficiency throughout the Sr efficiency calibration process by using our beta counters.

# Verification of $^{237}$Np activity concentration in a mixed radionuclides solution: recent experiences in chemical separation and measurement by alpha spectrometry

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A procedure was developed and tested for the measurement of $^{237}$Np activity concentration (1 – 10 Bq/g range) in a solution containing comparable activities of other actinides ($^{230}$Th, $^{234}$U, $^{238}$U, $^{238}$Pu, $^{240}$Pu and $^{241}$Am) as well as significantly greater activities from $^{60}$Co, $^{90}$Sr and $^{137}$Cs. Gamma and x-ray photon emissions of $^{237}$Np and its daughter $^{233}$Pa were not visible from this radionuclide mixture in a direct measurement with a high purity germanium gamma spectrometer. The unavailability of $^{236}$Np ($t_{1/2} = 1.54 \times 10^5$ y) spike precluded the application of isotope dilution mass spectrometry. Due to the absence of a suitable long-lived alpha-emitting tracer, gamma-emitting $^{239}$Np ($t_{1/2} = 2.36$ d) was used as the yield monitor after "milking" it from ca. 22 kBq of $^{243}$Am. Weighed aliquots of the freshly separated and purified $^{239}$Np solution were added to the sample solutions. One aliquot was prepared as a comparator in 5g of 2M $HNO_3$ solution in a sealed glass ampoule. After inducing radiochemical exchange between $^{237}$Np and $^{239}$Np in the sample solutions, co-precipitation and extraction chromatography techniques were used to chemically separate Np. The final sources for both gamma and alpha spectrometry were prepared by co-precipitation with 50 µg of Nd as $NdF_3$. Calibrations of (1) the gamma spectrometry system for $^{239}$Np relative geometry between ampoule and $NdF_3$ sources and (2) the alpha spectrometry system for the absolute geometry of $^{237}$Np-$NdF_3$ sources were carried out. Results indicated an average positive bias of 10% (5 determinations) with respect to the accepted $^{237}$Np activity concentration which was most likely attributable to incomplete separation from $^{230}$Th.

# Occurrence and Distribution of $^{210}$Pb and $^{210}$Po in California Groundwater Determined by a Single Sample Analysis Method

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## ABSTRACT

The overall goal of our study was to investigate the occurrence and distribution of naturally occurring radionuclides in California groundwater. To achieve this we sampled one hundred and twelve groundwater sources from all the major aquifers in the State. In the first part of our study we targeted $^{224}$Ra, $^{226}$Ra, $^{228}$Ra, total uranium, gross alpha and these results were published in the Journal of Health Physics, 89, 2005 (in press). The second part of the study, which is the subject of the current presentation, involved the measurement of $^{210}$Pb and $^{210}$Po. Of the wells sampled, fifty-one groundwater samples were selected as having the highest potential of dissolved $^{210}$Pb and eighteen samples as having dissolved $^{210}$Po for analyses.

Both radionuclides were separated from a single 5-L sample of groundwater, followed by alpha counting for $^{210}$Po and proportional counting for $^{210}$Pb. Polonium was quantitatively deposited on a nickel disc from strong hydrochloric acid solution using a modified HASL procedure. $^{209}$Po was used as a tracer to assess chemical recovery. After the $^{210}$Po separation, $^{210}$Pb was separated from the resulting solution using a modified method by Eichrom Industries, using a lead carrier to assess chemical recovery. Analysis of de-ionized water spiked with NIST-traceable $^{210}$Pb and $^{210}$Po standards demonstrated excellent accuracy and precision of these methods.

Groundwater wells that showed the highest concentrations of $^{210}$Pb and $^{210}$Po were distributed in southern California. The $^{210}$Pb activities ranged from 0.11 pCi $L^{-1}$ to 15 pCi $L^{-1}$ and the average and median $^{210}$Pb activities were 1.96 pCi $L^{-1}$ and 0.346 pCi $L^{-1}$ respectively. For both radionuclides more than 30% of wells were above the EPA's derived MCLs using current health risk models ($>1$ pCi $L^{-1}$ for $^{210}$Pb and $>0.7$ pCi $L^{-1}$ for $^{210}$Po). Based on these findings it is reasonable to conclude that lead and polonium can be a significant contributor to radioactivity in California groundwaters.

# Effect of Energy on Alpha-particle Counting Efficiency

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It is the long-held belief that alpha-particle counting efficiency does not depend on energy. Our goal is to test the belief that radionuclides counted at different energy levels will result in a different efficiency. If we are successful in providing evidence that defies this basic logic, it will revolutionize the need to obtain efficiencies for alpha radionuclides. In the process of defying this conventional wisdom, we also need to develop an effective method to accurately measure the peak area for alpha-emitting particles. Finding the best available software for alpha peak analysis will result in more precise Quality Control measurements, and establish more accurate standards for the future. Ten energy calibration sources of a mixture of Natural Uranium and Am-241 were created to be measured in 40 detectors. We analyzed the results using five methods from 3 different software (WinAlpha, Genie 2000, and Maestro). To determine the accuracy of our results, we took the ratio of the peak areas for Uranium 234 and Uranium 238 isotopes and compared them to that of the SRM Certificate. The WinAlpha Region of Interest (ROI) provides the most accurate results as it showed a 0.15% increase of the U234/U238 ratio from the NIST SRM certified value with a 1.53% uncertainty. Thus, we can conclude that energy has no effect on the alpha counting efficiency greater than 0.15% over 0.577 MeV.

# Centrifugal electrodissolution system for irradiated rhodium target material used for the cyclotron $^{103}$Pd production

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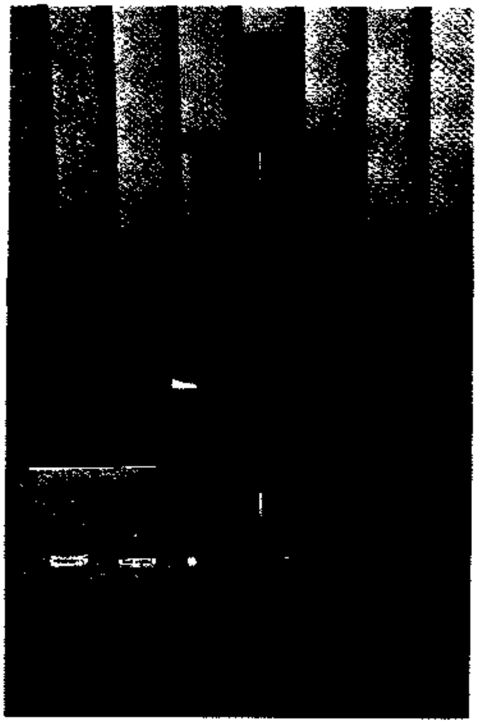
A new centrifugal electrodissolution technology was developed. It allows time-controlled electrodissolution of proton irradiated rhodium powder, fragments, cut wires and foils in a limited volume of hydrochloric acid.



The mini-reactor setup includes a hollow, cylindrical electrographite working-electrode (WE) receptacle fitted with an insulating PEEK bottom disk and with a PMMA cylindrical top-piece that increases the volume of the system. A cylindrical graphite counter electrode (CE) of appropriate diameter is mounted axially and extends up to 1mm of the bottom disk. The WE is mounted on the axis of a dc motor/tacho combination operating at 1000 rounds per minute. When filled with hydrochloric acid (40 ml, up to 12N) that contains the rhodium (up to 3 g) and when applying a high alternating current (up to 30 A) between working and counter electrode, centrifugal force and heat-generated fluid convection currents will result in the target material to be collected and to be rapidly dissolved at the lower section of the vertical wall of the WE where a current density up to 2 $A.cm^{-2}$ is obtained and where the concentration of the electroactive chlorine species dissolving the target material is maximum. The system is fitted with three sensor-feedback systems that allow volume (40 ± 2 ml), temperature (85 ± 2 °C) and current (30 ± 0.5 A) to be kept between narrow limits.

An extensive study of the parameters that determine the rhodium dissolution rate (dW(t)/dt in $mg.min^{-1}$) such as the input concentration of hydrochloric acid ($C_o$), the applied current (I) and the input weight of rhodium ($W_o$) in the reactor, resulted in a transcendental mathematical expression :

$$dW(t)/dt = -K.[1-e^{-b.C(t)}].[1-e^{-c.\Delta I}].[1-e^{-d.W(t)}] \qquad (1)$$

where: $W(t)$ = current weight of undissolved rhodium (mg)
$C(t) = Ceq.[1-e^{-a.t}] - C_o.e^{-a.t}$ = current concentration HCl in reactor ($mol.l^{-1}$)
$C_{eq} = C_o(1-k.I)$ $\Delta I = I - i_{co}$
K, a, b, c, d, k and $i_{co}$, experimental constants

A QB45 numeric integration algorithm based on Eq. (1) allows to calculate the time required to electrodissolve the rhodium up to a given solubilisation level (SL in g or %). Input data are $C_o$, I, $W_o$ and SL that acts as end-loop parameter of the algorithm. The output represents the dissolution rate and the remaining weight of undissolved target material as a function of time.

The maximum single-run capacity of the present system is limited to 3 grams of rhodium. The reproducibility of the system is excellent. For 10 runs ( with I = 25 A, $C_o$ = 12 N, V = 40 ml and T = 85 °C) each of 800 mg of rhodium fragments obtained from electroplated targets, a mean solubilisation level of 99.2 ± 0.3 % was obtained after the calculated dissolution time of 141 minutes for 99.0 % SL.

# Use of Thin Films to Control Recoil Contamination of Alpha Spectrometry Detectors

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**Introduction.** When an atom emits an alpha particle, the nucleus recoils with the same momentum as the alpha particle which has been emitted. The process is analogous to the familiar recoil produced when firing a rifle. The bullet is the alpha particle, and the rifle is the recoil nucleus. Under the low pressures used in alpha spectrometer chambers, the recoil nucleus has sufficient energy to reach the detector, where it is permanently embedded in the surface of the detector. If the recoil nucleus is also radioactive and decays by alpha emission, its characteristic alpha energy spectrum will be "seen" by the detector. Sill and Olson[1] have presented a thorough discussion of the phenomenon as well as a mechanical device to prevent detector contamination by recoil nuclei. They used a Cartesian diver to control the pressure within the counting chamber, leaving just enough air to stop the recoil nucleus with little effect on the energy of the emitted alpha particle, and with insignificant degradation of spectral resolution. Their approach, coupled with a reverse potential of ~ -6V to the source plate was highly effective in practically eliminating recoil contamination.

The best modern alpha spectrometry systems have built-in electronic pressure control devices that measure the actual chamber pressure. Control can be set by the user with amazing precision. Other systems have used a controlled leak as a means of allowing a very small amount of air to remain in the counting chamber. Older systems, however, do not have either feature, and regrettably, perhaps, some new systems were purchased without the anti-recoil option. For facilities that do not have recoil protection, thin films offer an excellent means of preventing recoil contamination. This presentation will show that by the use of thin films, control of recoil contamination can be achieved that is equal to or surpasses the pressure control/reverse bias techniques.

**How significant is recoil contamination?** A simple experiment can be used to demonstrate the recoil phenomenon. A sample containing 540 dpm of Th-229 was mounted for alpha spectrometry by $NdF_3$ micro-coprecipitation, using 50 µg of Nd carrier. A clean stainless steel plate was mounted in the top of an alpha spectrometry chamber in place of the detector. The Th-229 source was positioned beneath the plate as if it were the detector and the Th-229 was to be counted. The chamber was pumped to its normal pressure (<200 mTorr within two minutes, and then gradually decreasing to less than 20 mTorr). This defines the beginning of the exposure, which is analogous to the start of a count. The exposure was continued for 5 days in order to maximize the recoil contamination occurring on the stainless steel plate. At the end of the

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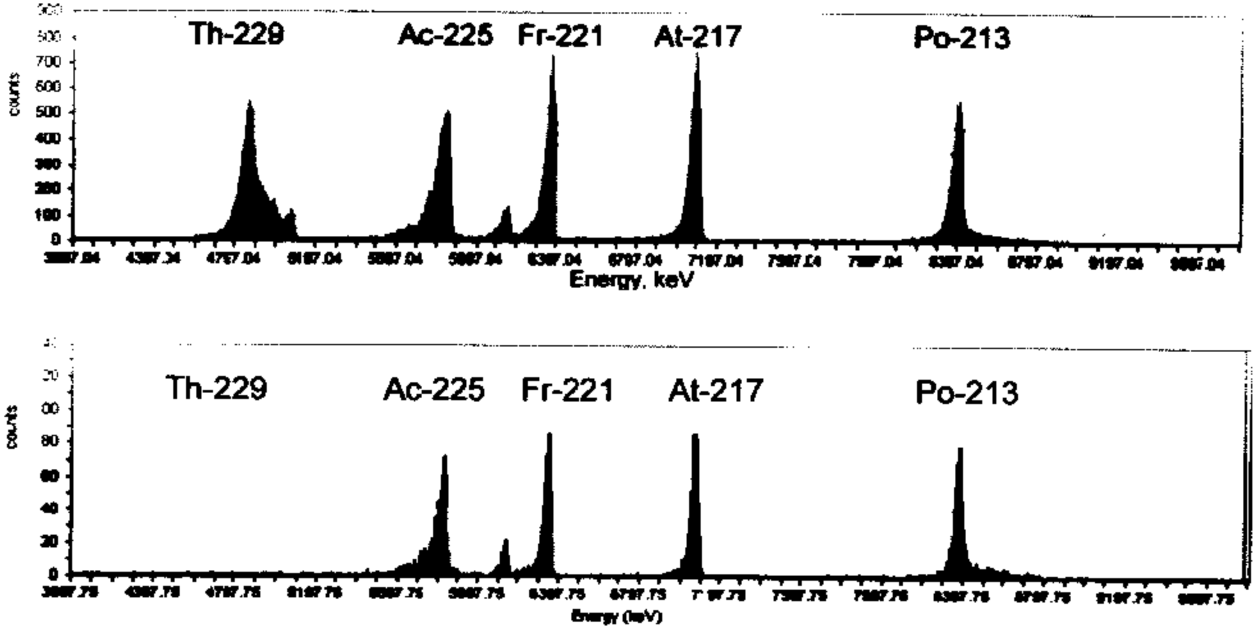
[1] Claude Sill and Dale Olson, *Analytical Chemistry 42*, 1596-1607 (1970)

exposure, the stainless steel plate was removed (and incidentally, covered with a thin film to prevent subsequent contamination of the alpha detector!) and the sample was counted for 48 hours (to generate enough counts in spectral ROIs for adequate demonstration). The startling results are shown in Figure 1.

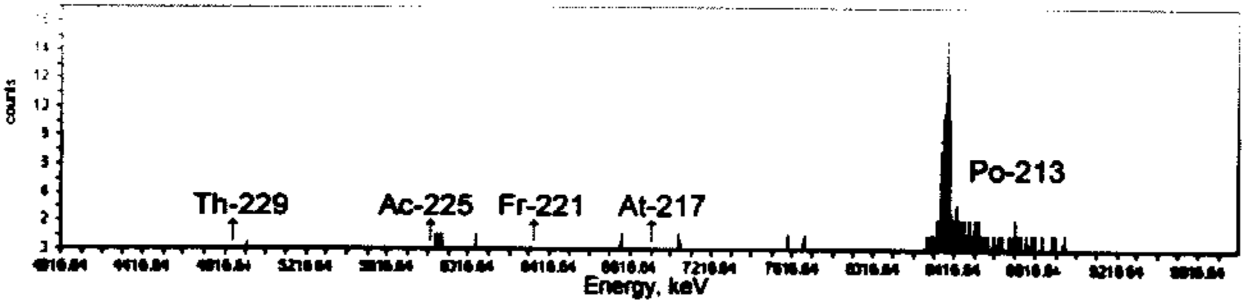
**Figure 1**



The peaks in the bottom spectrum are of Th-229 daughters. The first daughter of Th-229 is Ra-225, which is a beta emitter. The daughter of Ra-225 is Ac-225, and its presence in the spectrum indicates that Ra-225 is present on the stainless steel plate. Note also the superb resolution of the stainless steel plate. This is truly a massless source, producing resolution equal or better than what could be produced by electrodeposition. The recoil nuclei are permanently embedded in the stainless steel plate but the activity will decrease exponentially in accordance with the half-life of Ra-225. Herein lies the significance of danger associated with recoil contamination. If the recoil nuclei have short half-lives, the concern is minimal. However, the 1.91-year Th-228 daughter of U-232 makes uranium detectors the most often and most heavily contaminated by recoil.

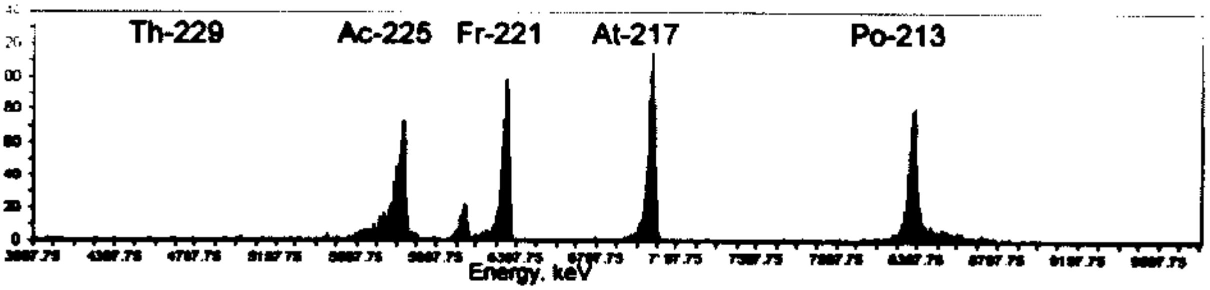
In contrast, look at Figure 2 to see what happens when the experiment is repeated with the Th-229 source covered by a thin film. In both experiments the exposure time was five days, yet with the source covered, the only significant peak is due to Po-213, whose half-life is less than a second. Polonium behaves like no other element with regard to recoil. Sill and Olsen discussed this at length. In their work, Po was not retained on the source plate even when reverse bias was

**Figure 1. Recoil plate exposed to Th-229 covered by thin film.**



applied. They found that only when a plate was heated was it possible to fix the Po and thereby control recoil. The well-known tendency of Po to spontaneously deposit onto Ag or Ni disks has resulted in a number of radiochemistry methods for determination of Po isotopes. It is therefore possible that Po may spontaneously deposit on any surface inside the spectrometer chamber, including sample holder, walls and detector. In the recoil experiments, if there is some time delay between exposure and counting of the exposed plate, we see "normal" Po-213 activity, i.e., the Po-213 activity is equal to its At-217 parent. This can be seen in Fig. 3 where some 9 days elapsed between exposure of the plate and start of count.

**Figure 3. Recoil plate 9 days after exposure to uncovered Th-229.**



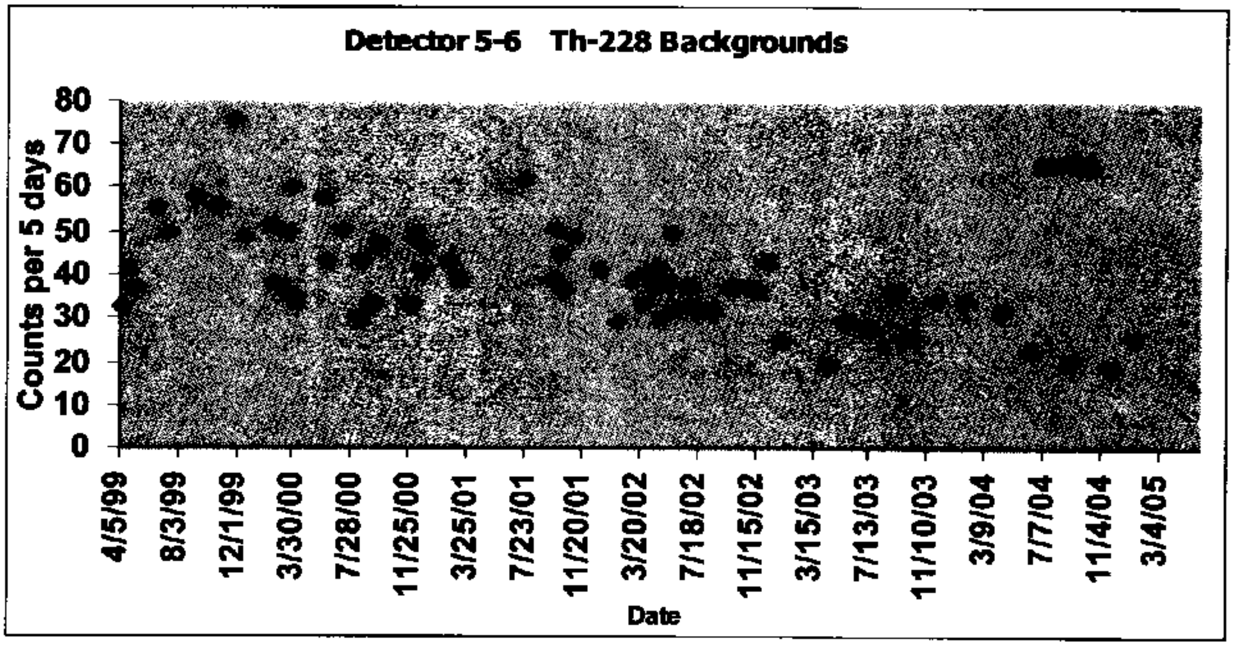
**<u>How do you stop a recoil nucleus</u>?** Sill and Olsen determined in their experiments that if the source to detector distance is 6 mm, an absorber thickness between 12 and 20 μg/cm$^2$ would stop recoil nuclei without significant impact on either the spectral peak position or resolution. We should be able to accomplish the same thing by applying a film of equivalent thickness directly to the source. The technique utilized at CEMRC produces films with an average mass of 100 μg. When applied to a 1-inch planchet (area 5.1 cm$^2$), the resulting absorber thickness is ~ 20 μg/cm$^2$.

It should also be pointed out that micro-coprecipitation may provide additional recoil protection not obtained with electroplated sources. This is due to the fact that 50 μg of Nd when precipitated as fluoride produces an additional 70 μg of absorber. The diameter of the $NdF_3$ precipitate produced by filtration equipment normally used for this purpose adds another 20 μg/cm$^2$ of absorber. Although not uniformly available to recoiling nuclei, it does offer an added measure of protection.

**<u>Preparation of thin films</u>.** The films are prepared by applying a drop of thin film solution to the surface of a large container of water. The surface area of the container must be large enough that the organic liquid can spread to its fullest extent without touching the sides of the container. As the drop of reagent spreads over the water, it cures to a "skin" in about 30 seconds. The film is then removed with a wire loop, dried, and applied to the sample. The thin film solution is prepared by mixing equal volumes of flexible collodion and iso-amyl acetate. It has been demonstrated at CEMRC that this reagent can be used for at least two months without significantly affecting the thickness of the films produced. With continual use, the cap of the vial is removed and replaced many times, and this allows for gradual evaporation of the solvents. As this occurs, the thickness of the resulting films will gradually increase over time. While this provides greater recoil protection, in time spectral degradation may occur.

**Results**. We have accumulated over five years of background data that shows conclusively that thin films can substantially reduce or eliminate the problem of recoil contamination. Figure 4 shows the Th-228 background of a detector used for counting uranium. Uranium-232 tracer is used, which means that Th-228 recoil would cause a gradual increase in the background in the Th-228 ROI. The data from this detector was selected for comparison because it is one of the worst cases and covers the greatest period of time. Obviously there has been significant contamination of this detector.

**Figure 4**



Our records do not indicate the exact date that we started using thin films. However, the chart clearly shows that the background on this detector is decreasing. Although the source of the original activity was very likely to be the result of recoil contamination, use of thin films has prevented further contamination.

**Conclusion**. Thin films represent a simple cost effective way to protect alpha spectrometry detectors from recoil contamination. No hazardous waste is generated in the preparation of thin films. The films can be easily removed should it become necessary to reprocess a sample. Although the focus of the data presented here is alpha spectrometry, thin films can also be applied to many other types of samples to afford a measure of stability during handling, counting, and storage.

# Massive Impact

## The Use of Collodion Films to Shield Solid State Alpha Detectors from Heavy Ion Recoils

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**Are impacts on your detectors by unwanted alpha-recoil ions costing you unnecessary money for repairs? If so, we have a cheap and easy solution that will save both your detectors and your money.**

Alpha-recoil contamination of solid state alpha spectroscopy systems leads to inaccurate measurements and wear on the detector. This contamination results in increased and variable backgrounds, larger measurement uncertainty, increased detection limit and eventual costly replacement of the device. Historically, in order to prevent recoil contamination, the use of $12 \mu g / cm^2$ air thickness and a six volt negative bias was used to control alpha recoil contamination (Sill and Olson, Analytical Chemistry, 42, 1596 (1970)). As an alternative, we have developed and tested thin collodion (polymer) films as a mechanism to improve the reduction of heavy recoil ion contamination. A nitrocellulose based collodion in iso-amyl acetate solution was prepared into a $12 \mu g / cm^2$ film and irradiated by a $^{238}$Pu source under a ten volt negative bias below a detector. Particle counts on the detector show a decrease in several orders of magnitude of recoil contamination when these films are used, even well after $10^9$ alpha-particles are detected. The mechanism by which these films stop recoil ions was shown to be impact energy loss, which then creates free carbon bonds in the nitrocellulose structure. Atomic Force Microscopy (AFM) was used to plot surface topography and impact damage on the films while infrared spectroscopic composition mapping was used to chart the chemical degradation of the film structure. Nanoindentation was used to plot changes in elasticity and modulus to form a mean lifetime for the films dependant on absorbed dose. It was found that thin single-use Collodion films do successfully shield solid state alpha detectors from wear and damage due to recoil contamination, improving detection limits and reducing the need for expensive replacements.

# Evaluation of Quench Corrected Liquid Scintillation Counting Instrument Blanks to Reduce False Positive Rate

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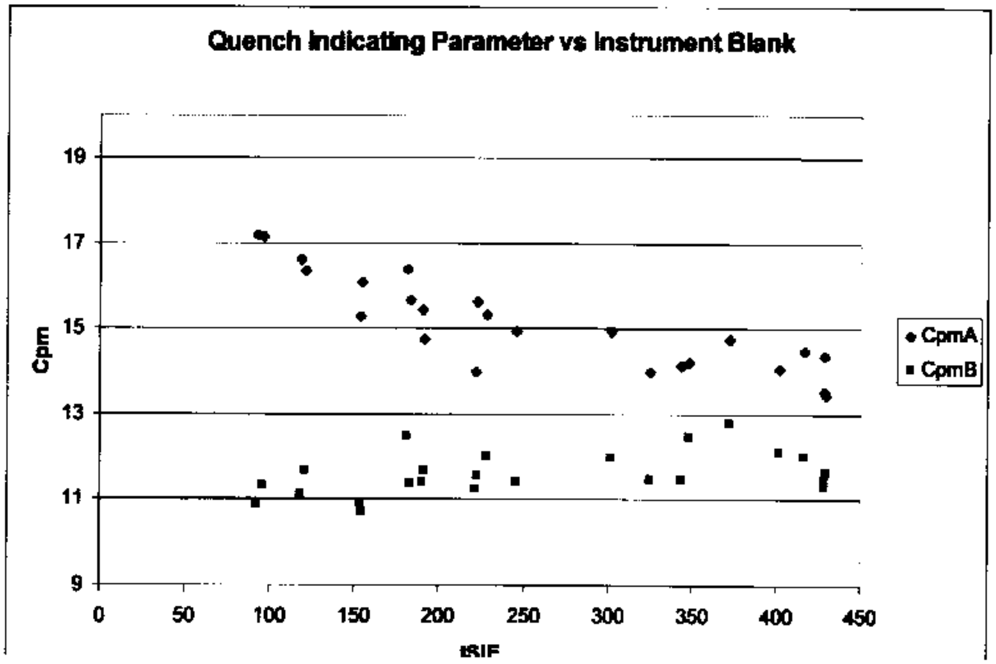
Tests were performed recently in this laboratory to evaluate the potential for quantitating quench corrected liquid scintillation counting instrument blanks. The purpose is to evaluate the use of these corrected values for adjusting sample counting data, thereby potentially reducing the false positive rate. Significant change in count from chemical quench was observed in the Tc-99 region of interest of 25 to 294 keV (CpmA). However, the count rate response was constant in the region of interest from 294 to 2000 keV (CpmB). See Attached Plot.



The background continuum is spilling higher energy counts into the region of interest with increased quench. Thereby the count rate is increasing instead of decreasing as one would normally expect. Further testing has simulated this effect by observing a similar count rate increase for a given window in the presence of a higher energy beta emitter. Similar testing is being performed on lower energy nuclides (e.g. Ni-63 and Pu-241)

This investigation is continuing and the various means of using quench corrected instrument blanks for adjusting sample concentration are being evaluated. It is desired to better match the instrument blank correction to the quench level of each individual sample. Since the background contribution for each sample is quench dependant. These corrections will be performed on historical data to determine the effect(s) on the possible false positive rate. This and newer data will be discussed during the oral presentation.

# The Radiological Assistance Program

Joel Swanson

The U.S. Government, through DOE and its predecessor agencies, has maintained a RAP since the late 1950s. RAP is designed to make DOE resources and monitoring and assessment assistance available to other Federal agencies, as well as state, local, and other governing agencies, for the explicit purpose of assisting in mitigation and evaluation of radiological incidents. RAP is implemented on a regional basis. This regional coordination is intended to foster a working relationship between DOE radiological response elements and those of state, local, other Federal agencies, and other governmental agencies that can be utilized to effectively resolve a radiological incident.

Requests for radiological assistance may originate from other DOE facilities; from Federal, state, or other governmental agencies; or from any private corporation or individual. Many of the requests will be handled by a telephone call or a written response. Other requests for assistance may involve radioactivity in serious accidents or possible hazards to the general public and may require the dispatch of trained personnel equipped with radiation monitoring instruments and related equipment necessary to evaluate and contain the hazard. Such assistance does not in any way preempt state, local, or other governmental authority or responsibility. DOE assistance for non-DOE incidents is limited to technical assistance, advice, measurement, and other resources as deemed necessary by the responsible agency authorities. All assistance will be coordinated through the appropriate state agency. Assistance is provided with the understanding that the duration of the commitment will depend on the nature and extent of the incident and the state and local resources available. RAP assistance will terminate as soon as the situation can be controlled by state, local, and/or commercial responders. The cost of the assistance will be the responsibility of DOE unless other agreements or reimbursement mechanisms exist.

# Using Existing Field Radiological Instrumentation to Influence Emergency Response Decisions

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In recent years, numerous emergency response organizations have acquired various types of instrumentation which may provide some indication of the presence of radioactive materials. In many cases, simple procedures can be employed with even the least-sophisticated equipment to give radiation protection professionals valuable decision-making information. This paper offers a "triage" approach to the interpretation of instrumentation responses in a radiological scenario.

The two most probable scenarios that would initiate a radiological response are as follows: a first responder (police, firefighter, emergency medical technician, etc.) has his radiation pager activated when responding to the scene of an accident or explosion, or a law enforcement officer (police, Customs/Border Protection, Port Authority, etc.) has his radiation pager activated when performing random or routine searches of people, places or things (e.g. cargo or other commerce). The first thing the emergency responder or law enforcement officer must recognize is that radiation is all around us, whether natural or man-made, and for radiation pagers to be useful they will alarm to these innocuous sources as well as to sources which pose a health threat to the responder/officer and the public.

Because of this non-discriminatory response, it is crucial that the wearer be given basic instruction on the operation and response of the radiation pager issued. Various radiation pagers have been purchased by federal, state and local emergency response and law enforcement organizations which have different capabilities and sensitivities. Most radiation pagers are sensitive to gamma radiation; some are also sensitive to neutron radiation. For gamma radiation most pagers use a NaI or CsI crystal or a Gieger-Muller (GM) tube. Some radiation pagers read out in dose rate, some in count rate, while others provide a relative intensity of the incoming radiation compared to background. The most common material for neutron detection is LiI or He-3 tubes. The readout of these simple radiation pagers for neutron radiation is typically in count rate or relative intensity.

Civil Support Teams and HAZMAT (federal or local) teams also often have portable survey equipment sensitive for alpha and beta radiation. Many federal, state and local emergency response teams are also being equipped with portable hand-held "Radio-Isotope Identification Devices" (RIIDs). The RIIDs may be equipped with NaI, CsI or HPGe crystals for isotope identification of gamma-emitting radioisotopes. Their readout may be in count rate or dose rate. Available options include neutron detection using LiI or He-3, a CZT detector (for better identification of low-energy gamma-emitters), a GM tube for monitoring high dose rate fields.

When a radiation pager alarms, several questions need to be answered quickly. These include: Is the alarm real? Where/what is the source of the alarm? Is it a problem? Is the source localized? What is it that caused the alarm? Do you need help? Each of these questions will be addressed in the following paragraphs.

The first question to be resolved is whether the radiation pager alarm was real on not. Most manufacturers indicate that their radiation pagers will have spurious alarms 2-3 times per day of normal usage. Some pagers are designed to periodically "chirp" (which sounds like the alarm) a few times per hour as a way to ensure the instrument is working properly. The quickest way to resolve an alarm is to back away so that the pager stops alarming and then re-approach the area where the alarm sounded. This tactic works when the radiation source is a fixed object. If the alarm does not repeat, it is possible that there was a source but it has moved away from your location. Often radiation pagers are worn clipped to the belt; this leads to a significant directional shielding by the body of the user. If the alarm is determined to be real, localization techniques can then be implemented.

The source of the alarm could be a person (carrying a medical treatment burden), a specific box, cargo container or vehicle, or it could be dispersed over a wide area. If localization is not possible, it could indicate a radioactive dispersal due to an accident. In this situation, the area should be cordoned off and additional support personal will most likely be required. If localization is possible then the determination of legitimacy may proceed.

While identification and localization of the source is being performed the responder/officer must also evaluate his own personal protection. Most organizations have prescribed dose limits or intensity limits for which their personal may safely work, and time limits for working in a known radiation field. If the radiation source exceeds operational limits the area should be cordoned off and additional support personal will most likely be required. If responding to an injury, the injured party should be moved away from the radiation source as quickly as practicable. Radiation intensity falls off very quickly with distance thus a short move could place the injured person and responders in a much safer position.

If the source of radiation is determined to be a specific container, vessel or vehicle, and the radiation levels are below operational suspension limits, the officer may determine the legitimacy of the source. Radioactive shipments must be marked according to national and international law, and be on the manifest of commercial shippers. In the US the Department of Transportation (DOT) regulates the movement of radioactive material; plackarding and allowable external dose limits are described in 49CFR173. Because the US no longer produces medical or industrial radionuclides they must be imported; medical isotopes are received primarily from Canada while industrial isotopes may come from Europe or Asia. A brief listing of common medical and industrial isotopes is shown in Table 1. Many other items found in routine commerce will contain naturally occurring radioactive material (NORM) at levels which may cause radiation pagers to alarm. A brief listing of items that may contain NORM is shown in Table 2.

**Table 1. Examples of Medical and Industrial Radioisotopes**

| Isotope | Typical Use | Radiation emitted |
|---|---|---|
| Cs-137 | Industrial (level gauges, moisture/density gauges, discrete sources) | Beta, gamma |
| Co-60 | Industrial (radiography sources, sterilization sources, discrete sources) | Beta, gamma |
| Am-241 | Industrial (smoke detectors) | Alpha, gamma |
| Tc-99m | Medical (stress tests) | Beta, gamma |
| I-125 | Medical (thyroid diagnostic) | Beta |
| I-131 | Medical (thyroid therapy) | Beta, gamma |
| Tl-201 | Medical (stress tests often in conjunction with Tc-99m) | Gamma |

**Table 2. Examples of NORM**

| Isotope | Typical Material | Radiation emitted |
|---|---|---|
| U-238 | Porcelain, tile, Fiesta Ware, ore | $\alpha$, $\beta$, weak $\gamma$ |
| Th-232 | Welding rods, optical lenses, engine parts | $\alpha$, $\beta$, weak $\gamma$ |
| Ra-226 | Luminescent devices and paints, fuel oils | $\alpha$, $\beta$, $\gamma$ |
| K-40 | Fertilizer, kitty litter | $\beta$, $\gamma$ |

Most of the medical isotopes emit low-energy gamma-rays thus are easily shielded and should not cause an excessive alarm on the radiation pager. If the dose rate or gamma intensity outside the shipping package is very high, a broken container might be suspected. Most NORM will emit radiation at increased levels but should not exceed any operational suspension limits. Significant radiation being emitted from a container of NORM should be investigated further.

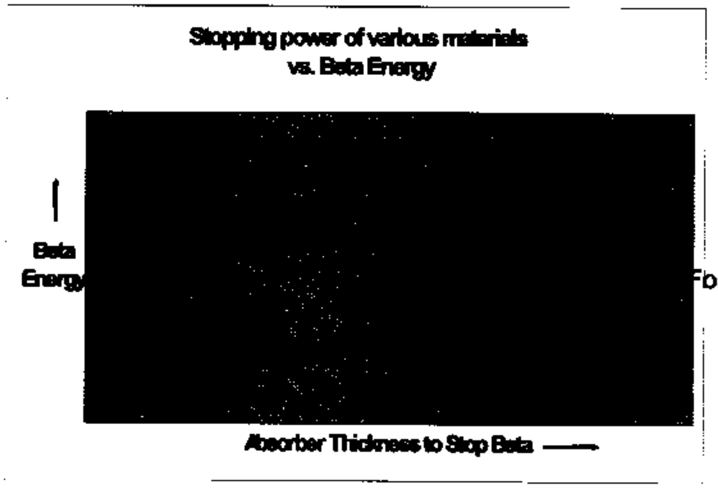
Neither medical isotopes nor NORM should emit neutrons however some legitimate industrial sources may emit neutrons. Well-logging and moisture gauges are the most common instruments to contain neutron emitting sources. These may be Cf-252, or more commonly "alpha-n" sources, consisting of Ra, Po or Am coupled with Be. These instruments must be on the manifest of the transportation vehicle. A repeatable, non-manifested neutron alarm on a radiation pager should trigger further investigation.

If the source of the elevated radiation is identified as a person, they may have had a recent medical therapeutic or diagnostic treatment. Experience has shown that persons with a medical burden may exhibit very high dose rates/intensity levels as measured by the radiation pager. It is not uncommon for a pager to alarm at several tens of feet from a person recently released from the hospital which performed the test or treatment. Depending on the isotope the person may be detectable for days to a few weeks. In some cases persons are not informed by their doctor of this side-effect. Pagers which use LiI for neutron detection may indicate the presence of neutrons when exposed to the gamma radiation emitted from persons with medical burdens. If possible, isotope identification should be performed to confirm the presence of the medical isotope. At a minimum, it should be confirmed that the radiation is emanating from the whole body, not a localized source, recognizing that the readings will be somewhat higher in the trunk due to the majority of body mass. Any inconsistencies should trigger further investigation.

Once a radiation pager alarm has been determined to be real, and the source of the radiation deemed to require additional support or investigation, some simple field tests may be performed to provide additional information to decision makers. If a RIID is available, it can be used to confirm the presence of medical, industrial or natural radioisotopes or of special nuclear material (SNM). Some RIIDs may indicate the presence of beta-emitting isotopes in the absence of gamma-emitters by observing the Brestraalung. This should be confirmed by other beta detection instrumentation.

If a RIID is not available, preliminary identification can be estimated using other tools available. For example, using a beta-gamma probe on a survey instrument, cover the probe window with your hand or a book. If the count rate drops to near background the radiation is predominately beta. The same conclusion can be drawn by turning the probe face away from the source; if the activity remains nearly constant it is predominately gamma radiation. Likewise, alpha activity will be reduced to near background when a piece of paper is placed between the source and the probe face. This technique is useful when a radioactive dispersion (spill or leak) is suspected. The approximate beta energy can be estimated by placing progressively thicker absorbers between the source and beta sensitive detector as shown in Figure 1. The average beta energy for some common radionuclides is shown in Table 3.

**Figure 1. Relationship Between Simple Absorbers and Beta Energy**



To verify neutron activity in the presence of a high gamma radiation field with a pager containing a LiI detector a few simple tests can be made. The neutron count rate should diminish more slowly with distance from the source than the gamma count rate, and the gamma activity should be more affected by shielding than the neutron activity. If the gamma count rate/dose rate is high and the neutron count rate drops significantly with distance from the source, the neutron reading is probably due to spillover of the high gamma radiation.

**Table 3. Average Beta Energy for Some Common Radionuclides**

| Nuclide | Average Energy (kev) |
|---|---|
| C-14 | 50 |
| Tc-99m | 85 |
| Ir-192 | 188 |
| I-131 | 192 |
| Sr-90 | 195 |
| Mo-99 | 443 |

Finally, the initial responder/officer must determine if there is a problem that requires additional assistance. Questions that need to be addressed include: does the RIID or other identification methods/processes agree with the isotopes listed on the manifest? Does the RIID confirm NORM where expected? Was the presence of neutron radiation confirmed? Use this and other law enforcement and/or emergency response training to sense problems. If the source of radiation cannot be confirmed as "innocent" secure the area and call for radiological response personnel, e.g. Radiological Assistance Program (RAP) team (DOE resource), HAZMAT, Civil Support Teams (CST). Refer to the Nuclear/Radiological Incident Annex of the National Response Plan as appropriate.

Based on the initial instrumental analysis the responder/officer can assist other responders and analytical laboratory personnel by providing the following information: (i) is it a fixed, localized radiation source or dispersed source? (ii) the relative amount and type of radiation, e.g. gamma, neutron, "mostly beta with a little gamma," "no significant alpha," "high energy beta," etc, (iii) if a RIID was used any nuclide identifications should be passed on to laboratory personnel, (iv) if transferable contamination is suspected this must be communicated to other personnel.

# Anatomy of an emergency exercise: EXFO.

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## INTRODUCTION

The Human Monitoring Laboratory (HML), which operates the Canadian National Calibration Reference Centre for *In Vivo* Monitoring, participated in an emergency exercise known as EXFO. The exercise was held at CFB Suffield (Alberta) from 21 Feb to 25 Feb, 2005. The HML team consisted of Gary Kramer, Ed Korpach, Renato Falcomer, and Jeff Cheung. The last three members were borrowed form other sections in the Radiation Surveillance and Hazards Assessment Division. The Director of the Radiation Protection Bureau, Jack Cornett, was the Health Canada Team Leader.

## THE EXERCISE

The exercise began with a briefing at 15:00 hrs on Day 1 (Tuesday 22 Feb 2005). The scenario presented was that there had been an explosion in a post office in the fictitious town of Wattopolis. Fire, police and hazmat personnel had responded to the incident. Emergency responders had transported injured persons to the local hospital and the area had been checked for biological, chemical and radiological presence. The latter was found a few hours after the incident had occurred. The hospital had reported persons were showing signs of radiation exposure: vomiting, nausea. The number of affected persons was unknown and a figure of twenty was estimated

The Province had requested Federal help and the duty officer of FNEP was initially informed. The Federal Nuclear Emergency Plan had been fully activated and various Federal Departments began to dispatch personnel and equipment to the town to commence operations at 08:00 on day 2.

Day 2: personnel from Health Canada, DRDC, CNSC, RCMP, NRCAN, and AECL arrived at the location designated as C&C, which was about 5 km north of the post office. It was decided that the aerial survey team should first fly over the area, followed by two ground teams. The first ground team to check the roads for alpha/beta contamination, and the second team to investigate the scene and assess the degree of radioactive contamination. The provincial medial officer needed isotope identifications as soon as possible so treatment for the injured persons could be refined. Within a few hours the number of affected persons was reduced from twenty to three or four. Ottawa was kept informed by a combination of telephone, and uploads to an FTP site.

Immediately after the first briefing the portal monitors were setup outside the C&C in tents. Power was a combination of generator and line. The final setup is shown in Figs. 1 - 2.

The roads around the post office were shown to be free of radioactive material and so was the area outside the post office. The fly-over identified a strong $^{137}$Cs signal with the presence of something else. Swipes taken at the scene showed it to be $^{82}$Br. Other nuclides were found in a nearby storage shed, but it was quickly resolved that this source were military supplies and not part of the exercise. The helicopter also flew the area around C&C and showed it to be free of any strong gamma emitters.

It was later revealed that two of the exposed people were terrorists and an address was found amongst their belongings. The helicopter was dispatched to survey the area, but it found nothing except some neutron sources belonging to the military for land mine detection. There was confusion for a while as it was not known if these sources were part of the exercise - it was resolved after about 30 minutes that the neutron sources were not part of the exercise. Meanwhile, a team was dispatched to survey the apartment. Not much was found here but papers pointed to another site.

During the day, the background levels around the portal monitors was measured and recorded. These measurements were performed to ensure that no radioactive contamination had entered the screening areas. The measurement is shown in Fig. 3.

A laboratory was found, which just happened to be in the location already checked by the helicopter and about 100 m from the C&C . Again two teams checked the area, one outside and the other inside. As for the post office, the most hazardous sources were isolated in shielding. As little as possible was disturbed to leave the area for police forensics.

At about 22:00 hrs, 4 of the 5 HC team members left to scene to get some rest. The remaining member manned to portals that were used to check incoming teams (in a real incident, they would have been used for the public). He remained on scene until ~05:00 hrs on day 3.

Day 3. A car accident occurred in the early morning. Police responded and radioactive source were found in the rear of the car. Also a city map with multiple locations marked with radiation symbols was found. All areas, except 2, were checked by the aerial team. This was also an exercise artifacts as no pilots in Canada, except the British currently located at CFB Suffield can fly ay 100 ft at night - so this would not be done in reality. Some areas were found to have radioactive sources, and some were found to be clean.

The 4 HC team members returned to the C&C at 07:30 hrs on Day 3 and two survey teams were checked by the portal monitors on their return. At 08:00 hrs the exercise was halted and the tents and other equipment were packed up (at 04:15 hrs FNEP had shut down).

# CONCLUSION

Problems encountered were generally of an artificial nature, but the exercise also highlighted some real problems. In no particular order:

- the teams' radios could not be used on the exercise ground by order of the military (artificial),
- the helicopter did not get proper clearance for the first flight so it was delayed (artificial),
- 4 out of 8 portal monitors were damaged in transit and did not work,
- some equipment was lost (jerry cans, extensions cables),
- the exercise did not simulate media interaction (artificial),
- the exercise did not simulate the effect of a panicked general public (artificial),
- the portal monitors were used in tents outside the C&C (probably artificial),
- one team member was arbitrarily eliminated from game play to simulate stress,
- a location flown over by the aerial survey team later became contaminated with a $^{60}$Co source - this should have been present all the time (artificial),
- team members could not drive around the exercise area with out an escort (artificial),
- the hospital was not simulated and the exposed personnel could not be questioned directly or monitored by team members (artificial),
- communications were a problem as the range of the supplied radios was too short,
- GR135s stopped communicating with their PC's for no good reason,
- the small computer network established by DRDC operated through a satellite phone so communication with graphics intensive applications (e-map) were very slow.
- the network could not be used if somebody used the satellite phone to make a phone call (NRCAN had other satellite phones in their inventory, but we were unaware of this at the time),
- teams did not know the resources of the other teams in detail,
- the portal monitors are sensitive to the cleanliness of supplied power,
- the RCMP robot was not exercised, although it was deployed,
- portal monitors were not exercised for high volume throughput.

Lessons learned from this exercise that need to be addressed so that HC can better respond in the future include:

- HC has insufficient personnel for an extended operation,
- crowd control equipment is absent from the HC equipment inventory,
- signs are absent from the HC equipment inventory,
- team members did not have sufficient communications equipment,

- toolkit for simple in-field repairs need to be present,
- dosimetry toolkit needs to be on HC laptop computers,
- laptop computers need to be wireless network enabled,
- equipment need to be packed better to resist the rigors of travel,
- laptops did not have sufficient tools (dose calculation etc) on board,
- departments should be aware of each others resources, ahead of time. A master list needs to be created.

# Improvement in Radioassay Emergency Response Capability: NRIP'05

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***Problem Statement:*** **Radiological emergency response decision making process will depend on a large number of rapid radioassay measurements. The quality of the decisions made will depend on the reliability of the radioassay results. The State and Federal radioassay emergency response laboratory community has never been exercised with traceable reference materials to evaluate accuracy, uncertainty and turnaround capability to identify shortfalls in capability and capacity. NRIP'04 was our first attempt to obtain information about the state of radioassay emergency preparedness with a limited number of participating laboratories. The information gained by the participant laboratories should be important to improve their capabilities and prepare a support quality system. The NRIP'05 radioassay emergency preparedness exercise was an examination of the improvement developed between the two exercises.**

***Response:*** **NIST has conducted two radioassay emergency preparedness exercises to evaluate the capability of a limited number of State, Federal and International radioassay laboratories:**

1. **Fluor Hanford, Richland, WA**
2. **Institute of Nuclear Energy Research, Taiwan**
3. **New Jersey Department of Health and Senior Services Environmental and Chemical Laboratory, Market and Warren Streets, Trenton, NJ**
4. **U.S. Department of Energy, Waste Isolation Pilot Plant Laboratories, Carlsbad, NM**
5. **U.S. Environmental Protection Agency, National Air and Radiation Environmental Laboratory, Montgomery, AL**
6. **Washington State Department of Public Health Laboratory, Shoreline, WA**

**The six NIST Radiochemistry Intercomparison Program participating laboratories received five replicate samples of spiked soil, water, air filter, urine, feces and their appropriate blanks then reported measurements of gamma-ray emitters, gross alpha, gross beta, and radioisotopics. The scope of the exercise was to evaluate the capacity of a laboratory to return accurate results within eight hours of sample receipt. Comparison of laboratory results with the NIST massic activity values were reported to each participant laboratory, and post-exercise conference calls for methodology comparisons were conducted for future response capability improvement.**

***Results and Impacts*:**

- **All laboratories met the eight hour turnaround time.**
- **Most of the reported measurements were for gamma-ray emitting radionuclides, some for gross alpha and gross beta analysis, and two laboratories reported radioisotopic results. Reported expanded uncertainties (k=2) were as follows: 3 - 100 percent for gamma-ray measurements, 1 - 18 percent for gross alpha measurements, 3 - 17 percent for gross beta measurements, and 14 - 21 percent for radioisotopic results.**
- **For NRIP'04 NIST and the participating laboratories, learned much about laboratory capabilities. Ways to improve both the efficiency and effectiveness of the exercise and radioanalytical methodologies were discovered. Problems encountered included arranging for airfreight shipping priority, weather delays, sample packaging, sample geometry, reporting format, and uncertainty evaluations.**

- NRIP'05 results indicated significant improvement in capability in some categories of measurement, but showed that continued efforts are needed in the remaining categories.
- Preparation, verification and distribution of traceable test samples were crucial for unambiguous interpretation of the reported measurements.
- Discussions of lessons learned are underway to improve the operation of future exercises.
- Sharing radioanalytical methodologies with each other and discussing the merits of the methodologies is very beneficial for laboratory capability improvement.
- The exercise proved invaluable for the participating laboratories to begin the process of improving their response capabilities.
- The results of the exercises are being published in the open literature, and circulated to interested emergency response agencies.

# 51st Annual Radiobioassay & Radiochemical Measurements Conference Poster Presentation Abstracts

# Technical Program Schedule - Poster Presentations

| Poster Number | Authors/Co-Authors | Affiliation | Title |
|---|---|---|---|
| 1 | Sandra K. Fisher and David P. Hickman | University of California, Lawrence Livermore National Laboratory | Nuclear Accident Dosimeters at the Lawrence Livermore National Laboratory |
| 2 | Benjamin J. Hicks | Severn Trent Laboratories | Determination of Carbon-14 from Reactor Graphite |
| 3 | Benjamin J. Hicks, Joel Kempema, Steven L. Howard, Chelsea Jarrell | Severn Trent Laboratories; SAIC | Ra-226 by Alpha Spectroscopy, a Comparison of Preparation and Separation Methods by Two Independent Laboratories |
| 4 | Johnson, B.E., Esser B.K., Hunt, J.R., Conrado, C.L. and Guthrie, E | University of California, Lawrence Livermore National Laboratory | High resolution radionuclide analysis of sediments for reconstructing local releases |
| 5 | Gottfried Kueppers | Operation Management Nuclear Infrastructure, Department Decontamination (B-ND), Germandy | Fast source preparation procedures for alpha-spectrometry of the actinides |
| 6 | Dominic Lariviere, Ana Paula Packer, Chunsheng Li, Jing Chen, Jack Cornett | Radiation Protection Bureau, Canada | Age dependence of natural uranium and thorium concentration in human bone |
| 7 | P. E. Cisneros, C. J. Duffy, K. M. Israel, D. L. Kottmann, C. A. Lance, D. M. Lopez, L. J. Miller, S. B. Ortiz, J. L. Roach, F. R. Roensch, J. A. Weeks, R. E. Steiner, and S. P. LaMont | Isotope and Nuclear Chemistry Group, Los Alamos National Laboratory | Simplified Sample Preparation Chemistry for the Analysis of Plutonium by Thermal Ionization Mass Spectrometry (TIMS) |
| 8 | Weijia Li, Chunsheng Li, Jiujiang Zhao, Stephen Kiser, Weihua Zhang, Dominic Lariviere, Jack Cornett | Radiation Protection Bureau, Health Canada; Carlton University | A Study toward Real Time Monitoring of Radionuclides in Fresh Waters Using Diffusive Gradients in Thin Films Technique Coupled with Germanium Gamma Ray Detector |

# Technical Program Schedule - Poster Presentations

| Poster Number | Authors/Co-Authors | Affiliation | Title |
|---|---|---|---|
| 9 | Yun-Gang Liu, Shiyamalie R. Ruberu and S. Kusum Perera | California Department of Health Services | Gross Alpha /Beta Analyses of Groundwater by Liquid Scintillation Counting |
| 10 | H. K. Meznarich, E.d. Perez, K. Iwatate, D.G. Farwick | Flour Hanford | Technicium-99 Analysis at the Waste Sampling and Characterization Facility |
| 11 | Joseph Mwakapumba | Nevada State Health Laboratory | Sample pre-treatment and dissolution methods for analysis of radium-226, radium-228, and radium-224 by Liquid Scintillation Counter |
| 12 | A.M.G. Pacheco, M.C. Freitas, A. Machado, S. Sarmento, M.S. Baptista, M.T. Vasconcelos, J.P. Cabral | Technical University of Lisbon; Technological and Nuclear Institute; University of Port; Portugal | Vanadium accumulation in tree bark and epiphytic lichens at three sectors of Portugal |
| 13 | Arthur Scott and Philip Panter | Ontario Ministry of Labour - Radiation Protection Monitoring Service | Ontario Nuclear Reactor Surveillance Program Tritium in Air |
| 14 | Donivan R. Porterfield, Lav Tandon, and Edward R. Gonzales | Los Alamos National Laboratory | Software Coincidence Spectrometry Using Commercially Available Instrumentation and Custom Developed Software |
| 15 | J. M. Rankin, G. F. Payne, and N.E. Bores | Oak Ridge National Laboratory | Intercomparison Studies Spikes Urine Samples used for Method Development and Validation |
| 16 | Bernd Kahn, Robert Rosson, Liz Thompson and Jeff Lahr | Georgia Tech Research Institute | Radioanalytical Chemistry |
| 17 | Donald E. Dry, Joseph W. Sullivan | Los Alamos National Laboratory | Advanced Analytical Data Handling for the Bioassay Program at LANL |

UCRL-ABS-213476

# Nuclear Accident Dosimeters at the Lawrence Livermore National Laboratory

Sandra K. Fisher, David P. Hickman, Ph.D.

The Lawrence Livermore National Laboratory (LLNL) is required by DOE Order 10 CFR 835.1304 to provide nuclear accident dosimetry in "installations possessing sufficient quantities of fissile material (that) potentially constitute a critical mass, such that the excessive exposure of individuals to radiation from a nuclear accident is possible." Certain facilities at LLNL (e.g., the Plutonium facility) meet the above criteria. At LLNL, the nuclear accident dosimeter (NAD) is incorporated into the Panasonic TLD holder. The NAD is comprised of neutron activation elements that allow the neutron fluence for various neutron energy regions to be determined. A dose estimate in rads is calculated by applying dose conversion factors. LLNL utilizes wall-mounted and fixed NADs, as well as personal NADs. This poster describes the components of the NADs used at LLNL and how they are processed.

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

# Determination of Carbon-14 from Reactor Graphite

Benjamin J. Hicks
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Given the task of quantifying the amount of carbon-14 present in reactor graphite, several chemical tests were run concurrently in order to determine which would be the most effective oxidizer of this extremely stable matrix. Only chemical methods were investigated, as Severn Trent St. Louis does not currently possess the equipment to thermally oxidize graphite. Of the many chemical schemes tried the most promising were a Friedel-Crafts reaction from the 1800s, a mixture of concentrated perchloric acid and fuming nitric acid, and Severn Trent St. Louis' standard reaction mixture.

Using the Friedel-Crafts reaction, graphite was converted to *mellitic acid* by submerging the graphite in potassium persulfate and placing it in the cold. Slowly, too slowly, beautiful needles of crystalline mellitic acid formed in the vials. They were highly radioactive containing both carbon-14 and tritium. The two isotopes were easily separated using liquid scintillation, but only partial oxidation had occurred, and it was impossible to determine the exact quantity of the sample that had been counted. One solution was to purify the mellitic acid, obtain a dry mass, and use this mass to calculate the amount of graphite originally oxidized. Unfortunately, this logic is flawed, unless it is assumed that the graphite sample is 100% carbon by mass. This is not true, but at least one report found on the Department of Energy's website lists the purity of German reactor graphite at 99.999 % carbon. So it would seem that this assumption is a fairly safe one. Ultimately, the chemistry of mellitic acid posed more problems than could be solved in the few weeks time frame Severn Trent was given for this project. Much of this failed attempt is included because there exists a potential to develop a procedure using this oxidation method. It is safe, easy, and has the ability to determine carbon-14 and tritium nearly simultaneously. This method will be pursued to completion in time.

The attempt to oxidize graphite using concentrated perchloric and fuming nitric acid was partially successful and holds some promise, especially if the mixture is heated and various metal catalysts are used. However, this approach was abandoned because it was not certain that the pace of the graphite oxidation could be controlled enough to prevent an explosion. The addition of heat did dramatically accelerate the reaction, but even if the graphite cannot chain react and explode. It was not known if any of the products of the reaction are themselves explosives. Given the safety concerns surrounding these attempts they were halted.

Finally, combinations of Severn Trent St. Louis' normal carbon-14 reaction mixture which includes potassium permanganate, potassium persulfate, and sulfuric acid were tested. The first attempts were complete failures, but cycling these oxidizers in a specific order begins to oxidize milligram amounts of graphite. The graphite matrix becomes more susceptible to oxidation with each successive cycle. The reactions were carried out in ordinary erlenmeyer flasks where the reaction mixture is heated to boiling. The flasks were connected to carbon dioxide traps via a water trap. The water trap controls tritium contamination, while the carbon dioxide trap forms calcium carbonate, thereby capturing carbon-14. Once enough calcium carbonate forms to completely cloud the tube (about four cycles), the precipitate is centrifuged and cleaned using multiple water washes and plated. This method, after only four cycles, or roughly an hour, still has a relatively low yield for graphite oxidation, perhaps five percent. Thus it is necessary to weigh the calcium carbonate and extrapolate back to determine how much graphite the mass on the planchette represents. Again, the assumption that the entire mass of the sample consists of carbon needed to be made. The percentage of sample mass other than carbon should be low enough to avoid any meaningful bias, but by no means is this assumption absolutely perfect.

At this point, a clean, but extremely radioactive, precipitate is dried and already on a planchette. Rather than attempt to transfer this precipitate to a scintillation vial, calibrations are performed on a gas flow proportional counter for carbon-14. The resulting mass attenuation curve is acceptable, however, much efficiency has been sacrificed in order to avoid scraping the precipitate off the planchette. It would be preferable to count by liquid scintillation not only for the efficiency, but also for the spectral information. However, the primary aspect altered by efficiency is the detection limit which becomes relatively inconsequential when the samples of interest are in the microCurie per gram level.

Lastly, this test method does not have a true control, as a graphite standard with a known amount of carbon-14 could not be procured. Until such time as this type of standard exists, an extremely important tool is missing.

In conclusion, given the unique difficulties inherent to the graphite matrix, and the unavailability of a graphite standard, quality testing can still be performed inexpensively for carbon-14 in a graphite matrix.

# Ra-226 by Alpha Spectroscopy, a Comparison of Preparation and Separation Methods by Two Independent Laboratories.

Benjamin J. Hicks, Joel Kempema, Severn Trent Laboratories; Steven L. Howard, Chelsea Jarrell, SAIC.

Accelerated clean up at DOE sites has demanded shorter turn around times for laboratory analytical results. This has generated interest for determining Ra-226 without having to wait for ingrowth and /or decay of daughter nuclides. Traditional methods for performing Ra-226 analyses require ingrowth periods or decay periods for daughter nuclides to accurately quantify Ra-226. New analysis methods being developed include alpha spectrometry, liquid scintillation counting and alpha scintillation utilizing a new instrument design.

At this time alpha spectroscopy has generated the most interest, but currently there is not a promulgated separation chemistry. A draft ASTM drinking water method using chromatography separation is being pursued by the ASTM D19.04 committee. Different from the ASTM study, which utilizes chromatography separation methods; this comparison will examine precipitation chemistries, by two independent laboratories in water samples and soil reference materials.

UCRL-ABS-215959

# High resolution radionuclide analysis of sediments for reconstructing local releases

Johnson, B.E., Esser B.K., Hunt, J.R., Conrado, C.L. and Guthrie, E.B

Radionuclides provide essential chronological and geochemical tracers in sediment systems. The Seaplane Lagoon at the former Alameda Naval Air Station on San Francisco Bay is an ideal site for demonstrating analytical techniques that quantify depositional trends of radionuclide laden sediments. Within the lagoon sediments, $^{137}$Cs from atmospheric weapons testing and excess $^{226}$Ra from the releases of luminescent paint facilities on the base are tracers measured by gamma counting at Lawrence Livermore National Laboratory. Twenty sediment cores 1 – 3 meters in length were collected from the 45 hectare lagoon in order to capture the spatial variability of sediment contamination. Cores were frozen on site and later sectioned into 1 to 2 cm intervals. Over 400 samples in total were analyzed in an iterative approach to identify the 1963 peak in $^{137}$Cs and profile excess $^{226}$Ra. The chronology of sediment accumulation established by $^{137}$Cs and $^{210}$Pb analysis in conjunction with information on site history is applied to historically reconstruct excess $^{226}$Ra for each of the twenty cores. For selected increments of historical time, 2-D Kriging estimates are made to quantify spatial concentrations at un-sampled locations and provide a basis for quantifying temporal fluxes to the lagoon from the nearby outfalls.

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

# FAST SOURCE PREPARATION PROCEDURES FOR ALPHA-SPECTROMETRY OF THE ACTINIDES

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A very fast and simple source preparation procedure for the $\alpha$-spectrometric determination of the actinides has been developed. It is based on two liquid-liquid extractions and direct evaporation of the extractants - containing the actinides isotopes - on the counting dishes. From the sample two thin-layer counting sources are prepared to take into account the $\alpha$-spectrometric interference of the nuclides $^{238}$Pu - $^{241}$Am and the poorly resolved $^{228}$Th - $^{241}$Am, where one counting source contain the Th-, U-, Np- and Pu-isotopes and the other one the Pa- and Am-isotopes. Except for Pa, the chemical yields are in the range of 97 and 99 % so that no yield tracers are necessary. Starting with the dissolved sample, the two counting sources can be prepared in about 30 minutes. This preparation procedure is preferably applied to medium-active samples from the nuclear power plant waste stream, but can also be used for low-level environmental samples after suitable pre-concentration procedures.

## Introduction

Important constituents of nuclear power plant waste are the long-lived $\alpha$-emitting nuclides of the major actinide elements, Th, Pa, U, Np, Pu and Am. Their identification and determination in radioactive waste and environmental samples is important for waste management and - because of their radiotoxicity - for environmental monitoring.

The methods widely used for the measurement of the activities of $\alpha$-emitting nuclides, i.e. the preparation of thin-layer counting sources for $\alpha$-spectrometry, are complicated and time-consuming. Because of the rather poor reproducibility of complicated procedures in respect to the chemical yields the addition of yield tracers is necessary. Furthermore, most source preparation methods described in literature do not take into account the interference of the nuclides $^{228}$Th – $^{241}$Am and $^{238}$Pu - $^{241}$Am in the $\alpha$-spectrum.

The aim of this work was to develop a fast and simple source preparation method with high chemical yields and good reproducibility. The problem of interference was taken into account by the preparation of two counting sources for each sample whereby the interfering nuclides are separated.

## Experimental

The separation of the elements Th, Pa, U, Np, Pu and Am from the sample matrix is based on two liquid-liquid extractions.
In a first step the elements thorium, uranium, neptunium and plutonium are extracted from a 0 - 3 M nitric acid sample after addition of $Al(NO_3)_3 \cdot 9\,H_2O$ with a solution of 1 % methyl trioctylammonium chloride in methyl isobutyl ketone (MIBK), while protactinium and americium remain in the aqueous phase. After increasing $Al(NO_3)_3 \cdot 9\,H_2O$ concentration in the aqueous phase protactinium and americium are extracted with a solution of 10 % methyl trioctylammonium chloride in MIBK. Aliquots of the organic phases are then evaporated on counting dishes of stainless steel and 5 cm in diameter. After evaporation the dishes are heated intensely over a Bunsen burner to remove traces of remaining solid organic impurities (Fig. 1). With this procedure high chemical yields of 97 to 98 % could be achieved for the investigated elements. Because of the high chemical yields and the high reproducibility of ± 1 %, no addition of yield tracers is necessary (Fig. 2 and Tab. 1).
For α-spectrometry the "Alpha Analyst" from CANBERRA with passivated, implanted, planar silicon (PIPS) detectors was applied.
A typical α-spectrum (ash sample) is shown in Fig.3

1 ml dissolved sample (0 - 3 M $HNO_3$)

+ 5 ml $Al(NO_3)_3$ solution (500 g $Al(NO_3)_3 \cdot 9\,H_2O$ + 32 ml ammonia per liter)

+ 0.05 ml 0.1 M $KMnO_4$ solution

After a few minutes extraction with 5 ml of a solution of
1 Vol.% methyl trioctylammonium chloride in MIBK.

0.5 ml of the organic phase is evaporated on a counting dish.

(**Source A**; for **Th-**, **U-**, **Np-** and **Pu-**isotopes)

↓

Aqueous phase
+ 5 g $Al(NO_3)_3 \cdot 9\,H_2O$

After dissolution extraction with 5 ml of a solution of
10 Vol.% methyl trioctylammonium chloride in MIBK.

0.5 ml of the organic phase is evaporated on a counting dish.

(**Source B**; for **Pa-** and **Am-**isotopes)

*Fig. 1.* Source preparation procedure

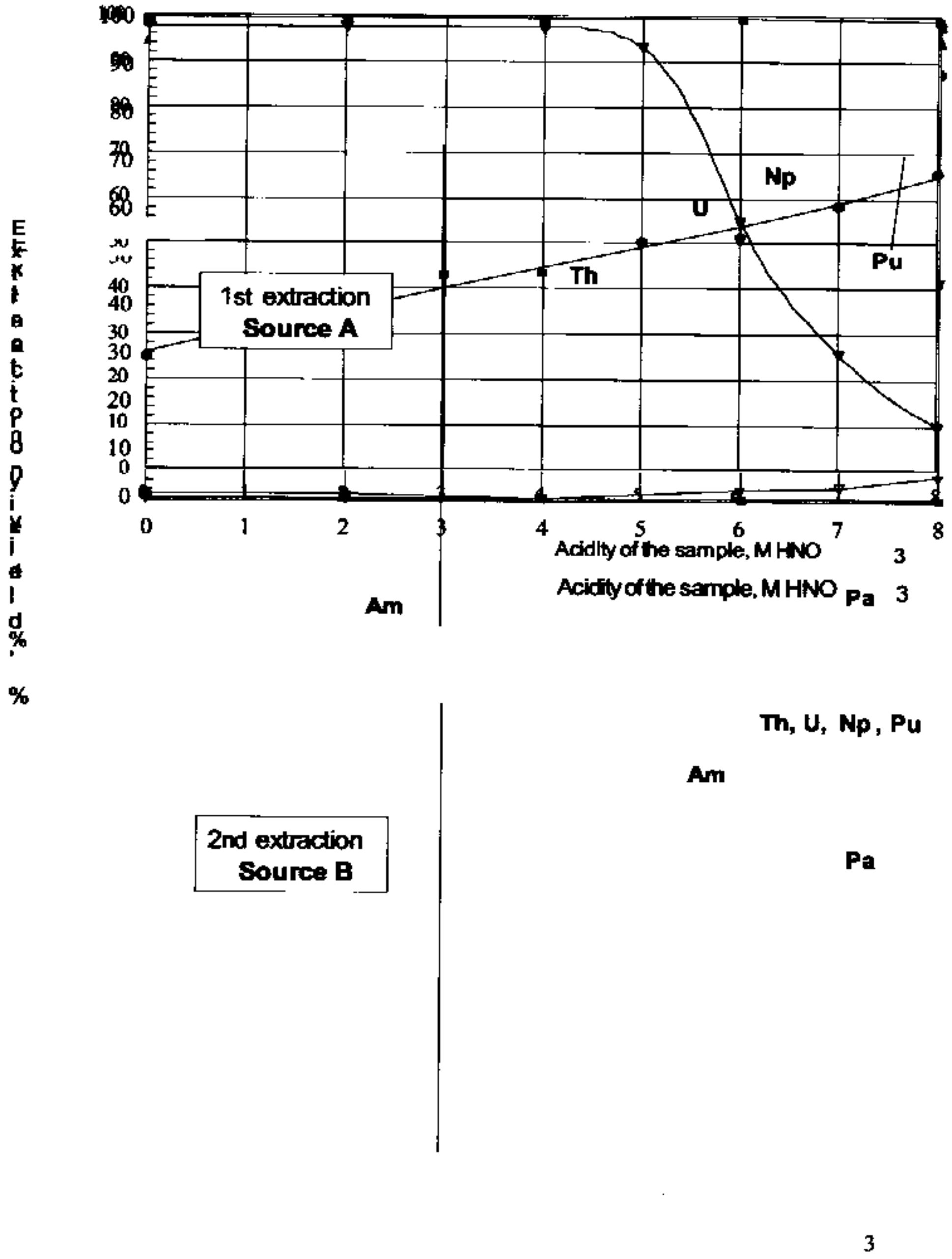

*Fig. 2.* Extraction yields as a function of the nitric acid concentration in the sample

| Element | Chemical yield, % | Source |
|---|---|---|
| **Th** | 99.0 ± 1.0 | A |
| **U** | 99.1 ± 1.0 | A |
| **Np** | 97.8 ± 1.0 | A |
| **Pu** | 99.5 ± 0.5 | A |
| **Pa** | 36.0 ± 2.0 | B |
| **Am** | 98.0 ± 1.0 | B |

*Table 1.* Chemical yields for a sample acidity of 2 M $HNO_3$

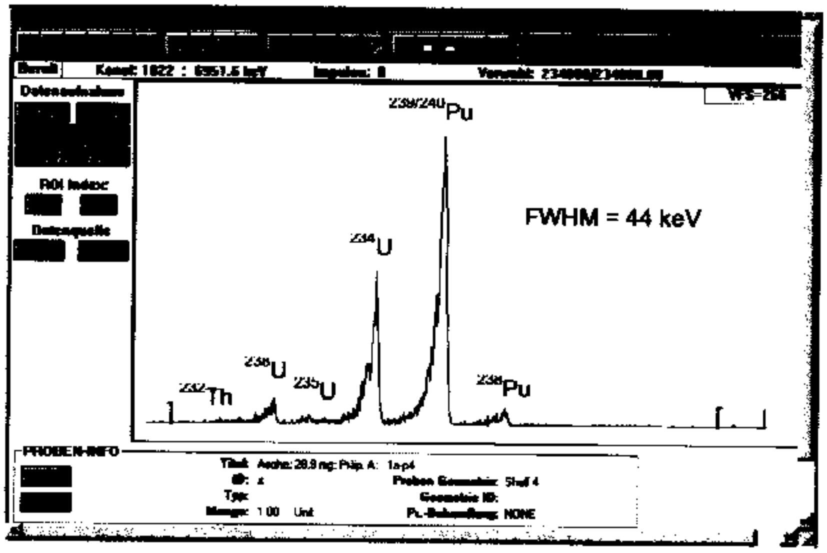

*Fig. 3.* $\alpha$-spectrum (source A) of an ash sample from the Juelich burning facility for radioactive waste

## Characterization of the source preparation procedure

- Very fast (about 30 minutes preparation time; starting from the dissolved sample)
- High chemical yields (97.8 – 99.5 %) and reproducibilities (± 1-2 %) for the elements Th, U, Np, Pu and Am.
- No addition of yield tracers.
- Discrimination between the interfering nuclides $^{228}$Th - $^{241}$Am and $^{238}$Pu - $^{241}$Am
- Quantitative separation of alkaline and alkaline earth elements (salt load) and of important long-lived fission and activation products such as $^{90}$Sr, $^{134}$Cs, $^{137}$Cs and $^{60}$Co.
- Similar energy resolution in comparison with "ideal sources" prepared by electrolysis.

## Literature

G. Küppers

"Fast Source Preparation for Alpha-spectrometry of Uranium and Transuranium Isotopes"

J. Radioanal. Nucl. Chem., 230, No. 1-2 (1998) 167

# AGE DEPENDENCE OF NATURAL URANIUM AND THORIUM CONCENTRATION IN HUMAN BONE

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**Abstract** – Uranium ($^{234,235,238}U$) and thorium ($^{232}Th$) isotopes are present at trace levels in the environmental resulting from their primordial origin and long half-lifes. As a result of their omnipresence, they enter our foodchain, where, they get eventually deposited in bones. For this reason, the study of U and Th concentration in bone is a good indicator of environmental radioactivity exposure. However, it is unclear at this point, if the concentration of U and Th increases, stabilizes, or decreases with age. To answer this question, we used the bone ash archive available at the Radiation Protection Bureau collected in the 1960-1970's. These bones were collected originally to monitor the impact of nuclear explosion on Canadian population but now represent a unique opportunity to study environmental radioactivity. Ash samples from various age groups and from two locations (Regina, SK and Winnipeg, MB) were digested using microwave digestion and $HNO_3$ and then analyzed by inductively coupled plasma mass spectrometry (ICP-MS) to determine the concentration of uranium, thorium, and trace metals. Thallium was used as an internal standard to evaluate the non-spectral interferences (matrix effect). Certified reference material (NIST-4356) was analyzed to evaluate accuracy and precision of the digestion protocol and ICP-MS measurements. The results of this study are important to validate if the retrospective internal irradiation dose calculation, performed using the International Commission on Radiological Protection (ICRP) age-specific biokinetic model, are accurate and realistic. In addition, this study will provide information concerning the levels of uranium and thorium in bone for Canadian population.

# Simplified Sample Preparation Chemistry for the Analysis of Plutonium by Thermal Ionization Mass Spectrometry (TIMS)

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Thermal ionization mass spectrometry (TIMS) is one of the few instrumental analysis methods capable of detecting plutonium in urine at concentrations that would be excreted by an individual with an 100 mRem, 50 year CEDE inhalation intake of Type S material. Los Alamos National Laboratory analyzes approximately 1800 urine bioassay samples per year by TIMS for plutonium as part of a comprehensive worker protection program. Prior to TIMS analysis, plutonium is radiochemically separated from the urine, electrodeposited onto a stainless steel planchet, and $^{238}$Pu and $^{239+240}$Pu are determined by alpha spectrometry. To prepare the sample for TIMS analysis, the plutonium is dissolved off of the planchet and radiochemically purified prior to electrodepostion onto a rhenium TIMS filament. Traditionally, plutonium was removed by leaching the planchet in 8M $HNO_3$ overnight, and then purified using a 1.0 mL anion exchange column followed by a 0.5 mL anion exchange column. This process took an entire day to complete (not including the leach step) and required the sample to be completely dried between the two columns to change the matrix. A new, rapid 15 minute leach in 8 M $HNO_3$ to remove the plutonium from the planchet followed by a single 1.0 mL anion exchange column that takes 4 hours to run was studied to replace the overnight leach and two column method. Both the overnight leach and 15 minute warm leach methods were tested in combination with two different anion exchange resins, AG MP1 50 – 100 mesh $Cl^-$ form, and AG MP1 100 – 200 mesh $Cl^-$ form, resulting in four possible combinations of parameters. Results from the four tests were compared to samples processed using the traditional overnight leach and two anion exchange column method. The best results were obtained with the overnight leach in 8 M $HNO_3$ and the AG MP1 100 – 200 mesh $Cl^-$ form resin, which produced TIMS data of comparable quality to the original method in approximately half of the time.

# A Study toward Real Time Monitoring of Radionuclides in Fresh Waters Using Diffusive Gradients in Thin Films Technique Coupled with Germanium Gamma Ray Detector

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**Abstract:**

Contamination levels of radionuclides in the Great Lakes region and other natural waters in Canada have attracted much attention over the decades due to the existence of nuclear energy facilities, mining activities and waste management stations there [1, 2]. To ensure environment safety and water quality, there is a need for continuous monitoring of radionuclides in waters.

This work presents a first water monitoring sensor system for radionuclides in waters, focusing on reliability, sensitivity, *in situ or* on site and real time measurement. This sensor system was composed of two techniques, diffusive gradients in thin films (DGT) [3, 4] and Germanium radiation detection [5, 6].

The DGT technique consists of three layers: the top layer is a layer of filter membrane to protect the second layer, a diffusive layer, from particles, and a bottom layer, a binding layer to serve as a sinker to kinetically collect the analyte species diffused through the diffusive layer. The analyte concentration, ***C***, in water can be determined by measuring the mass, ***M***, accumulated in the binding layer over certain deployment time, ***t***, after elution in an acidic solution according to DGT equation which is derived from Fick's first law of diffusion [3, 4]:

$$C = M \Delta g / D A t$$

Where $\Delta g$ and $A$ are the thickness and exposure area of the diffusive layer respectively, $D$ diffusion coefficient of analyte species in the diffusion layer.

To meet the specific requirement of uranium detection with DGT, a series of binding layers, Chelex 100, Uteva resin embedded in polyacrylamide hydrogel, and a commercially available Whatman P 81 ion exchange membrane were systematically investigated for DGT uranium application in functions of time, pH, ionic strength, analyte concentration. The experiments have confirmed that Chelex 100 and P 81 binding layer are applicable for DGT in fresh water pH and ionic strength ranges. Binding of uranium to Uteva resin was limited by the condition of certain nitrate concentration in sample solution. A traditional DGT diffusive layer, polyacrylamide hydrogel, and Whatman 1 Chr membrane were also studied under different ionic strength

conditions, and proved to be inert to uranium analyte, which is basic requirement for DGT.

To monitor trace uranium in fresh water (typically 0.7 ppb in Lake Ontario[7]) in real time, the best features of DGT, automatic kinetic preconcentration of analyte from water and *in situ* time averaged concentration measurements, are combined with sensitive Germanium radiation detector.

Germanium radiation detector was tested to detect the amount of radioactive uranium accumulated in the binding layer for its direct and sensitive functions, and compared with ICP-MS measurements. It has been shown that the detector is able to detect 100 ppb uranium in laboratory solutions with tradition DGT devices after 24 h deployment. The sensoring system, DGT technique modified with 754 cm$^2$ exposure area coupled with Germanium radiation detector, is then deployed in Lake Ontario and is able to detect 0.5 ppb uranium.

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# Gross Alpha /Beta Analyses of Groundwater by Liquid Scintillation Counting

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The method of gross alpha/beta analyses of water by liquid scintillation counting(LSC) previously reported from this lab (Wong et al., J. Radioanal. Nucl. Chem., 264: 357, 2005) is a viable alternative to the traditional proportional counting technique. The LSC procedure was validated by analyzing performance testing samples from EPA, ERA, DOE-QAP, and New York State in this study. Data showed this method is especially promising for water samples with high total dissolved solids (TDS). For gross alpha analysis by proportional counting, the maximum residual solid on the counting planchet is 5 mg/cm$^2$. As a result, small sample aliquots are used with high TDS waters, which give high detection limits. The liquid scintillation counting method can handle samples containing up to 500 mg dissolved solids and therefore can achieve much lower detection limits due to larger sample volume.

As a logical extension of that work, we have analyzed twenty California groundwater samples for gross alpha/beta using the liquid scintillation counting procedure and proportional counting. These samples covered a wide range of total dissolved solids (TDS) from 270 mg/L to 1560 mg/L. The activities measured by LSC were compared with those obtained from the proportional counting method. Gross alpha activities determined by proportional counting ranged from <3 pCi/L to >600 pCi/L. Using LSC method with 100 mL sample volume and 200 minutes counting time in our lab, a MDA of 2.5 pCi/L was achieved. The linear correlation between the two methods is excellent, with a correlation coefficient of 0.995. In the current study, data of 20 groundwater samples showed good agreement between the LSC method and the standard procedure with proportional counting. These results demonstrate that LSC method works well in the gross alpha analysis for the groundwater water samples.

## Technetium-99 Analysis at the Waste Sampling and Characterization Facility

H.K. Meznarich, Ph.D., E.D. Perez, K. Iwatate, and D.G. Farwick. Fluor Hanford, P.O. Box 1000, S3-30, Richland, WA 99352.

This presentation describes the method for analyzing Technetium-99 ($^{99}$Tc) that is used by the Hanford On-Site Laboratory, the Waste Sampling and Characterization Facility (WSCF). Performance of $^{99}$Tc analysis is also included in this presentation.

This method can determine $^{99}$Tc levels in water and soil matrices. Soil samples are prepared by leaching prior to chemical separation. Table 1 summarizes typical sample-size, types of preparation, and typical aliquot volumes used for chemical separation.

Technetium-99 is extracted from soil samples using 1M nitric acid and hydrogen peroxide or acidified by concentrated nitric acid and hydrogen peroxide for water samples, then separated from other beta emitters by either Eichrom Technologies TEVA$^{TM}$ Rad discs or TEVA$^{TM}$ resin columns. The rad discs or resins retaining $^{99}$Tc are then counted by a Liquid Scintillation Counter for $^{99}$Tc activity. The region of interest on the scintillation counter is set at 6 – 300 keV. The counting efficiency of $^{99}$Tc in each different media (e.g., TEVA$^{TM}$ resin column or TEVA$^{TM}$ Rad disc) is determined. Chemical recovery is performed for soil samples and is determined by with or without spiking $^{99}$Tc in each soil sample. Preparation blanks and laboratory control samples (LCS) are included in each preparation batch.

Table 1: Summary of Sample Preparation in Water and Soil Matrices

| **Matrix** | **Sample-Size** | **Preparation** | **Final Volume** | **Aliquot Used for Separation** |
|---|---|---|---|---|
| Soil by Rad Disc | 5gm | Acid Leaching: 30 - 50 ml 1 M $HNO_3$, 80°C for 2 hrs, 5 ml 30% $H_2O_2$, 80°C until clear | 200 ml with 0.01M $HNO_3$ | Apply 200 ml |
| Soil by Resin | 5 gm | Acid Leaching: 30 – 50 ml 1 M $HNO_3$, 80°C for 2 hrs, 5 ml 30% $H_2O_2$, 80°C until clear | No further dilution | Apply all to resin column |
| Water by Rad Disc or by Resin | 800 ml | 5 ml concentrated $HNO_3$ and 10 ml 30% $H_2O_2$, 90°C for 1 hr, cool and filter if necessary | All | Apply all |

$^{99}$Technetium can be calculated using the following equations:

- $E_{Tc\text{-}99}$: Counting efficiency for $^{99}$Tc using Counting Efficiency Sample by Liquid Scintillation Counting

  $E_{Tc\text{-}99}$ = (Counting Efficiency Sample Count Rate, cpm) – (Background Count Rate, cpm) / Spiked $^{99}$Tc Activity in Counting Efficiency Sample, dpm

- $R_{Tc\text{-}99}$: Chemical Recovery of $^{99}$Tc in each soil sample (SOIL ONLY)
- SS: Sample Size, L or g

  $^{99}$Tc (pCi/g or pCi/L) = (Sample Count Rate, cpm) – (Background Count Rate, cpm) / ($E_{Tc\text{-}99}$ * $R_{Tc\text{-}99}$ * 2.22 * SS)

The mean and standard error of mean (SEM) of the LCS recovery for water samples from January 2005 to October 6, 2005 are 102.7% ± 0.6%, respectively. The mean and SEM for preparation blanks in water are -0.38 pCi/L ± 0.22 pCi/L.

WSCF started to participate in the Mixed Analyte Performance Evaluation Program (MAPEP) water sample for $^{99}$Tc analysis in 2005. Several studies are presented for addressing issues associated with analyzing MAPEP water samples and evaluation of the newly developed $^{99}$Tc analysis in soil.

The mean and SEM for the percent recoveries of WSCF values against the known MAPEP water (MAPEP-MaW11) value and spiked MAPEP Soil PE samples are listed in table 2:

Table 2: WSCF Reported Values Compared Against Known PE Value or Recovery of Spiked Standard in MAPEP Soil Samples

| | **Water** | **Spiked Soil,** **TEVA Resin Column** | **Spiked Soil,** **TEVA Rad Disc** | **Spiked Soil,** **TEVA Rad Disc** |
|---|---|---|---|---|
| **Mean (%)** | 101.2 | 98.6 | 94.0 | 94.6 |
| **SEM (%)** | 1.4 | 1.7 | 1.4 | 2.0 |
| **N** | 10 | 4 | 8 | 4 |

The MAPEP water study shows that no acidification and a further dilution of MAPEP water samples are required. Recovery results for spiked MAPEP soil are acceptable for $^{99}$Tc analysis in soil. The $^{99}$Tc analysis for soil analyzed in MAPEP sample that included $^{99}$Tc is also underway.

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University of Nevada (Reno)

**SUMMARY:**

The following is presented:

(1) Sample pre-treatment and dissolution methods for analysis of radium-226, radium-228, and radium-224 by Liquid Scintillation Counter.

(2) The associated Liquid Scintillation Spectra in mineral oil and DIN scintillation cocktails.

(3) Relative merits and de-merits of the methods.

**PROCEDURE:**

A 1 liter sample volume of water is reduced to about 200 – 250 ml by evaporation and 1 ml of a 19 mg/ml solution of $Ba(NO_3)_2$ is then added. The solution is cooled and the pH adjusted to a value of 2-3. Radium isotopes (together with lead-210) are then co-precipitated with Barium as sulfates by adding 6 ml of 1 M $H_2SO_4$ to the boiling solution. The precipitate is repeatedly centrifuged and washed with de-ionized water until neutral.

(A) Radium-226

Dissolve the sulfate precipitate from above in 10 ml of 0.25 M of alkaline EDTA and heat in a water bath for complete dissolution. Cool and then transfer to a scintillation vial to which has been added 5 ml of mineral oil cocktail. Complete the transfer by washing with a total of 5 ml de-ionized water. Shake to mix and store for 3 – 4 weeks for radon to in-grow. Count on the liquid scintillation counter by normal or alpha-beta mode.

(B) Radium-226, Radium-228 and Radium-224 simultaneously

To the sulfate precipitate from above, add 10 ml of 0.136g/ml solution of sodium acetate ($CH_3COONa.3H_2O$) and 10 ml of a saturated solution of potassium carbonate ($K_2CO_3$). Digest the mixture by gently boiling and stirring for at least half an hour. Maintain the total volume above 20 ml by replacing the evaporated water with de-ionized water. The sulfate will be converted to a carbonate while the lead-210 forms an acetate complex and is taken up in the supernate. Centrifuge and wash the carbonate precipitate three times and then dissolve in 0.5 – 1 ml 2M $HNO_3$ acid. Transfer the solution to a scintillation vial to which has been added 19 ml of a DIN cocktail. Complete the transfer by washing with an additional 0.5 ml of 2M $HNO_3$ acid. Shake to mix. Let sit in the dark for 1 hour to dark adapt and for chemiluminescence to fade away. Count on the liquid scintillation counter in the alpha-beta mode.

# Vanadium accumulation in tree bark and epiphytic lichens at three sectors of Portugal

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Within an overall project of comparing native biomonitors with transplanted ones, *Platanus hybrida* bark and *Parmelia caperata* thalli were concurrently exposed from November 2003 to September 2004, for periods of 2 months. Lichen transplants were kept upon their substrates, and total (wet + dry) deposition was collected according to the same schedule. Experiments were carried out at three sites close to the western Atlantic coast – Viana do Castelo (just Viana, hereinafter), Sacavém and Sines – and located in the northern, central and southern regions of mainland Portugal, respectively. Discontinuous and continuous modes of exposure were set up through the experiments. Biological monitors were assessed by $k_0$-standardized INAA, and total-deposition samples by ICP-MS.

Wet-deposition volumes were as follows: Viana > Sacavém > Sines, except in the period January-March, for which a similar order of magnitude has been found at all sampling locations. Given this, enhanced elemental release would be expected at Viana. Dry deposition was virtually null at Sines and Sacavém in the period May-July, and, generally speaking, its extent (mass) has been within the same order of magnitude for every site. Exception was an amount for Viana in November-January that largely exceeded what was observed for the other two locations. Vanadium availability in total deposition was similar for Viana and Sines, and much lower for Sacavém. As observed for cobalt [1], highest levels of vanadium in the exposed bark and lichen tissues were reached at Sines, likely due to an association of elemental availability and alternate wet/dry periods. At Viana, even if nominal availability was the highest, the larger wet deposition may as well have hindered the retention of vanadium. Conversely, Sacavém featured the lowest vanadium availability in total deposition, but since there was again an alternation of wet/dry terms, the bioaccumulation of vanadium in both bark and epiphytes yielded biological concentrations similar to those observed up north at Viana.

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# Ontario Nuclear Reactor Surveillance Program – Tritium in Air

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The Province of Ontario contains three nuclear power complexes holding 20 reactors, and a research complex with 3 research/medical isotope production reactors. The Fermi II nuclear power reactor in Michigan is within 15 km (10 miles) of the Provincial border. Radiation Protection Monitoring Service (RPMS) is part of the Ministry of Labour and carries out an independent Nuclear Reactor Surveillance Program.

Canadian power reactors are moderated and cooled with heavy water ($D_2O$). Neutron capture produces tritiated heavy water (TDO). Leakage of heavy water leads to rapid isotopic exchange with water vapour in the air, so the reactors release airborne tritium in the form of HTO.

The Reactor Surveillance Program has used essentially the same methods to measure airborne tritium concentrations around the power reactor complexes since its organization in 1985. Atmospheric water vapour is collected over 4-week intervals in 100 g silica gel cells near population centres around the three nuclear power reactor complexes. The water is removed from the gel by heating, and the tritium concentration measured by liquid scintillation beta counting. This value is corrected for dilution by isotopic exchange with the residual and bound water in the silica gel to give the tritium concentration in atmospheric water vapour.

This presentation illustrates the equipment employed, the correction equation theory, and the procedure used to measure the parameters.

# Software Coincidence Spectrometry Using Commercially Available Instrumentation and Custom Developed Software

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We will report on our on-going work to examine the potential for software coincidence counting in the analysis of actinide nuclear materials. We are most interested in the use of commercially available counting systems that provide easy access to list mode time stamped data, e.g. Ortec digiBASE and DSpec Pro multi-channel analyzers.

While gamma-gamma coincidence spectrometry has many advantages we are also examining the potential for alpha-gamma coincidence spectrometry given the photon poor decay schemes of many actinide isotopes.

The commercial availability of counting systems providing list mode time stamped data can allow the easy acquisition of the data. However, the lack of software for the flexible analysis of the resulting data is a definite challenge in making use of software coincidence counting for routine analyses.

LA-UR-05-3147

# INTERCOMPARISON STUDIES SPIKED URINE SAMPLES USED FOR METHOD DEVELOPMENT AND VALIDATION

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## Abstract

Oak Ridge National Laboratory's (ORNL) Intercomparison Studies Program (ISP) provides customers with natural urine samples spiked with known activity levels of radioisotopes. These samples are commonly used for quality control purposes in routine analytical operations. ISP also provides spiked urine samples for purposes such as method development and validation. Forty single-blind natural uranium spiked urine samples were supplied to ORNL's Radiobioassay Laboratory for use in the development of an improved method for the determination of $^{235}$U and $^{238}$U in urine by inductively coupled plasma-mass spectrometry. When the results from this sample set were compared to the actual spike activities, a small negative relative bias was found for thirty-nine of the samples. Based on their past performances and their confidence in the ISP spike sample preparations, the Radiobioassay Laboratory re-examined the method; and further changes were made to the procedure. The Radiobioassay Laboratory then analyzed an additional twenty uranium spiked urine samples using the modified procedure and found that the accuracy of the data set significantly improved.

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# Radioanalytical Chemistry

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This textbook is intended as a guide to the functioning radioanalytical chemistry laboratory. It presents a 1-semester course on the senior or graduate level to train students to work in, manage, or interact with a laboratory devoted to radionuclide measurement. A functioning radioanalytical chemistry laboratory has two components: the wet laboratory to prepare samples for measuring radionuclides, and the counting room to perform the measurements. The samples being considered here are primarily for environmental monitoring in media such as air, water, vegetation, foodstuff, wildlife, soil, and sediment, along with other sample types which may be used for bioassay or process control.

Radioanalytical chemistry is the division of the discipline of nuclear and radiochemistry devoted to analyzing samples for their radionuclide content. The specialty of radioanalytical chemistry utilizes the combined strategy of identifying radionuclides in a sample matrix and purifying the radionuclide(s) of interest by chemical methods, then measuring their disintegration rate ("activity") by nuclear methods. First developed by Mme. Curie and those who continued her work, radioanalytical chemistry became a field of inquiry at the turn of the twentieth century. Early efforts were devoted to purification and elemental characterization of the radionuclides in the naturally-occurring uranium and thorium decay chains. Later, other terrestrial and cosmic-ray-produced radionuclides were identified by these techniques. The number of known and characterized radionuclides increased dramatically with the development and application of nuclear-particle accelerators in the 1930's and nuclear-fission reactors in the 1940's.

In the 1950's and 1960's, nuclear chemists and radiochemists were trained at many schools to participate in nuclear studies and radiological monitoring. Since then, the number of pertinent degree programs and the students who graduate from them has declined markedly. In addition, most of the practitioners who contributed to the flourishing of radioanalytical chemistry from the 1940's to the 1960's no longer are active, due to retirement and/or advancing age.

Given the scope of the fields requiring radiochemistry and the shrinkage in number of practicing radiochemists, it has become increasingly apparent to those in the field that there are fewer radioanalytical chemists available than will be needed if present trends continue. Writing this book was thus invested with some urgency by the sponsoring agency, NNSA-DOE. Chapter authors and reviewers were sought who were or continue to be active contributors to the development of radioanalytical chemistry.

This textbook was written to provide a guide for students and new graduates in applying the skills of radioanalytical chemistry to the needs that exist today. Second, it serves to preserve and transmit the practical aspects of the specialty, gleaned by longtime practitioners of the discipline.

The guidance contained in the text should serve for all of radioanalytical chemistry, including various research and application uses. From the seventeen included chapters, the student should attain the knowledge necessary to skillfully perform the following duties:

- Operate the radioanalytical chemistry laboratory safely and in accord with regulations, with assured quality and cost-effectiveness;
- Plan identification and measurement of radionuclides through a combination of chemical separation and radiation detection methods;
- Select sample size, sample processing, radiation detection instruments and peripherals, and measurement period to match detection sensitivity specifications;
- Identify radionuclides by their chemical behavior and radiation, and resolve ambiguities by expanding or revising separations and measurements;
- Calculate radionuclide concentration values and their uncertainty with consideration of the radionuclide decay scheme, sample processing losses, radiation detection efficiency, and interfering radiation.

This text is intended to accompany a class that functions as part of a program. The program should include at least prerequisite courses in (1) analytical chemistry lecture and laboratory and (2) nuclear physics and radiation detection laboratory.

The professional who is being trained in this field must be familiar with nuclear physics applied to radionuclide identification and measurement. As indicated in the list of chapters in Table 1, this topic is introduced in Chapter 2 to refresh the reader's memory. Detailed information pertinent to the main radiation detectors routinely used in the radioanalytical chemistry counting room is given in Chapter 8. Chapter 9 discusses radionuclide identification by decay scheme and gives examples.

Analytical chemistry applied to sample preparation and radionuclide separation is presented in Chapter 3 with general information on the purification processes in the wet-laboratory that underlie radioanalytical chemistry. Information specifically associated with the behavior of radionuclides in aspects such as their low concentration and the effect of radiation emission is given in Chapter 4. A portion of Chapter 5 describes the usual samples that are submitted to the radioanalytical chemistry laboratory to indicate the sample matrix that must be treated and the context in which analytical results are applied.

The chemistry work in the radioanalytical laboratory is described in chapters 5, 6, and 7. Chapter 5 discusses initial preparation such as dissolution or concentration before analysis. Chapter 6 discusses the radioanalytical chemistry process and gives numerous examples of separation methods. Chapter 7 describes preparation of the counting source to be submitted for the measurements described in Chapter 8.

The next four chapters are devoted to various aspects of data interpretation and presentation, and quality assurance. Chapter 9 considers interpretation of data for radionuclide identification. Chapter 10 reviews the important topics of data calculation, measurement uncertainty, and reviewing and reporting the results. Chapter 11 describes the quality assurance plan that must govern all laboratory operations. Chapter 12 discusses methods diagnostics to correct analytical and measurement problems that can be expected to plague every laboratory. Although the order of these groups was designed to maintain learning continuity, for laboratory practice, some of the contents of these chapters must be considered at the very beginning.

The following two chapters address laboratory operation. Chapter 13 describes laboratory design and management in terms of a model radioanalytical chemistry laboratory and its staffing, maintenance requirements and costs of operation. Chapter 14 discusses the practice of laboratory safety and the management elements that reinforce safety.

The last three chapters consider special aspects of radioanalytical chemistry. Chapter 15 describes various automated systems that are used to measure radionuclides in the counting room and in the environment. Chapter 16 is devoted to identification and measurement of radionuclides beyond the actinides. These are research projects at the cutting edge of radiochemistry that apply novel rapid separations and measurements of a few radioactive atoms from which much must be inferred. In Chapter 17, several versions of mass spectrometers combined with sample preparation devices are described. The mass spectrometer has been used in the past to detect a small number of radioactive atoms per sample, but the instrumentation and methodology have so improved in recent years that MS now serves as a reliable alternative to radiation detection for radionuclides with multiple-year half lives.

The textbook is intended to cover every skill set that the radioanalytical chemist will need to become a competent practitioner. It is arranged to effect an easy presentation for the instructor. Its appendices are a valuable reference for further research by both the interested student and the seasoned chemist. We present the text with confidence that it will prove an asset in training the next generation of chemists in this important discipline.

The text editor is Bernd Kahn. The preparation was overseen by and Editorial Advisory Board that consists of the chapter authors listed with asterisks in Table 1 and also Richard Perkins, Isabel Fisenne ( resigned ) and Jake Sedlet (deceased). The text will be published by Kluwer (now Springer). An accompanying Laboratory Manual is being prepared by Moses Attrep and Bernd Kahn. The preparation of the text book and laboratory manual is being supported under DOE/NNSA Grant # DE-FG07-01ID14224.

### *Table 1 Chapters and Authors*

1. Introduction, B. Kahn *
2. Radiation Detection Principles, M. Attrep* (LANL retired)
3. Analytical Chemistry Principles, J. Lahr* and B. Kahn*
4. Radioanalytical Chemistry Principles, J. Lahr*, B. Kahn*, and S. Morton* (INL retired)
5. Sample Collection and Preparation, R. Rosson*
6. Applied Radioanalytical Chemistry, B. Kahn*, R. Rosson*, L. Thompson*
7. Preparation for Sample Measurement, B. Kahn*
8. Applied Radiation Measurements, J. Keller* (ORNL)
9. Radionuclide Identification, B. Kahn*
10. Data Analysis and Reporting, K. McCroan (EPA) and J. Keller*(ORNL)
11. Quality Assurance, K. Inn* (NIST), L. Thompson*, P. Greenlaw (DHS), and L. Selvig
12. Methods Diagnostics, B. Kahn*
13. Laboratory Design and Management, C. Porter* (EPA retired ) and G. Murphy* (UGA)
14. Laboratory Safety, G. Murphy*, A Wickman (GTRI) and P. Schlumper (GTRI)
15. Automated Radionuclide Analysis Systems, H. Miley (PNNL) and C. Aalseth (PNNL)
16. Chemistry Beyond the Actinides, D. Hoffman* (LBNL)
17. Mass Spectrometric Radionuclide Analysis, J. Wacker*(PNNL), C. Eiden (PNNL), and A. Lehn (PNNL)

# Advanced Analytical Data Handling for the Bioassay Program at LANL

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LA-UR-05-4975

Technical Area: Instrumentation, Software, and Automation Tools
Presentation type: Poster

## Abstract

The LANL Bioassay Program generates data from a variety of sources including benchtop measurements, alpha spectrometry, and thermal ionization mass spectrometry (TIMS). Future plans call for expanding our capabilities which will require integrating data also from liquid scintillation (LS) counting and inductively coupled plasma-mass spectrometry (ICP-MS) analysis. Currently, analytical data is processed by individual analysts and uploaded by automated routines to a central LIMS. Paper copies of all data are then routed for approvals, and archived offsite at a federal repository. We are currently in the process designing a system that will help streamline this process. This new system will automatically collect and upload analytical data from each type of counting system to a MS SQL Server. Each analytical data packet will then enter into an automated workflow process where it will be analyzed, approved, and sent to a reporting tool. After each step in the process, digital signatures will be captured. This workflow process will reduce data entry errors and help expedite the sample management process. Each counting system will have its own .NET module written in VB. The workflow process will have a web-based interface programmed using .aspx and XML. An email and pager notification system will also be used to notify analysts of completed or pending samples. Crystal Reports will be used to print up any paper copies of the data that are needed. This system will run on a MS Server 2003 platform and will use IIS and MS SQL.

# Notes

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# RRMC 2005

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