HADMÉRNÖK

VII. Évfolyam 1. szám - 2012. március

Ferencz Bernadette - Vágföldi Zoltán
ferenczb@npp.hu - vagfoldi@hotmail.com

NATO 2ND SAMPLING AND IDENTIFICATION OF RADIOLOGICAL AGENTS LABORATORY PROFICIENCY TEST RESULTS (SIRA-2008)

Absztrakt/Abstract

Az alább cikkünkben áttekintést adunk az Észak-atlanti Szerződés tagállamainak 2008. évi radiológiai összemérési gyakorlatáról (2008 NATO Sampling and Identification of Radiological Agents II. Laboratory Exercise), ismertetjük annak célját és a mérési eredményeket, a megszerzett tapasztalatokat. Az összemérési gyakorlaton 13 NATO tagország 14 honvédségi/védelmi céllal fenntartott laboratórium vett részt – a szervező „Institute of Technology La Maranosa" Spanyol laboratóriumot is beleértve - a NATO/LG7/SIBCRA munkacsoport égisze alatt. Magyarországot a Magyar Honvédség Radiológiai Laboratóriuma képviselte, szakmai tanácsadóként és a mérések elvégzésében a Paksi Atomerőmű Zrt. és a Mezőgazdasági Szakigazgatási Hivatal Központ Radioanalitikai Referencia Laboratóriuma is részt vett.

This article demonstrates the results and experiences of the 2008 radiological laboratory exercise of the Party States of the 2nd North Atlantic Treaty Organization Sampling and Identification of Radiological Agents (NATO SIRA-2008) laboratory exercise. On this measurement exercise 14 defence dedicated laboratories of 13 NATO member countries – including the organizing institute "Institute of Technology La Maranosa" (ITM, Madrid, Spain) – took part, with the support of the NATO/LG7[1]/SIBCRA[2] working group. The Hungary was represented by the Hungarian Defence Forces Radiological Laboratory, with the Paks Nuclear Power Plant Radioanalytical Laboratory and the Agricultural Management Bureau Centre's Reference Laboratory for Radioanalysis acting as technical advisor and providing control measurements.

Kulcsszavak/Keywords: *SIRA gyakorlat, radionuklid azonosítás, gamma-spektrometria, alfa- spektrometria ~ SIRA exercise, radionuclide identification, gamma spectrometry, alpha spectrometry*

[1] LG/7: Land Group 7 on Joint NBC Defence (NATO workgroup)
[2] SIBCRA: Sampling and Identification of Biological, Chemical, and Radiological Agents (standardized procedure for sampling and identification procedures of CBRN agents according to AEP-66 NATO Handbook)

INTRODUCTION

The aim of this SIRA laboratory exercise was to compare the preparedness and capabilities of the NATO member countries' laboratories, preparing them for a possible "live" radiological or nuclear emergency or threat. The other aim of the comparison was also to prove the preparedness and performance of the participating laboratories (presented in Table 1) in the case of a given examination or measurement. This exercise continues the aim of 1st SIRA exercise. [1] It was based on a radiological transport accident and the analysis of an unknown long term stable radioactive liquid sample. A SIRA[3] is a NATO description of in situ survey, sampling and analysis. [2] After receiving the sample the laboratories have to report the description of radio analytical method, detected radioisotopes and their concentrations. According to the final result the laboratories give advice to incident commander or decision maker to manage hazardous situation.

RESULTS

A radiological scenario was simulated, and some laboratories have demonstrated being ready to produce a quick and accurate answer and radio analysis result. The scenario was a crash by a plane carrying a thermonuclear bomb, with no nuclear explosion occurring, but the bomb produced contamination when tactical explosives dispersed the radioactive charge. The scenario was a simulation of a real situation that happened in the past. A selection of radioisotopes from those that were present in the bomb, was made according to principal pollutants were present in a real case after the crash. In the Table 1 we present the participating laboratories.

N°	**Country**	**Participating Laboratories**	**Short name**
1.	Canada	Defence Research&Development	Def. R&D
2.	Czech Republic	NBC Defence Institute	NBC DI
3.	France	SPRA	SPRA
4.	Germany	Deployable NBC Analytical Laboratory	Deployable NBC AL
5.	Hungary	HDF Radiological Laboratory	HDF RL
6.	Italy	Centro Tecnico Logistico Interforze (Nuclear, Biological, Chemical)	CT Logistical Interforce NBC
7.	Netherlands	Organization for the Prohibition of Chemical Weapons	OPCW Laboratory
8.	Norway	Norwegian Defence Research Establishment FFI	Def. Res. Estab. FFI
9.	Poland	Military Institute of Chemistry and Radiometry	MI of Chem. and Radiometry
10.	Slovakia	RCBO Reference Chemical and Radiological Laboratory	Ref. Chem. and Rad. Lab.
11.	Spain (organizer)	Institut Technologico "La Maranosa"	ITM
12.	Sweden	Swedish Defence Research Agency	FOI
13.	United Kingdom	Atomic Weapons Establishment	AWE
14.	United Kingdom	Defence Science and Technology Lab.	DSTL

1. table. List of participating laboratories

The task of the participants was to identify radionuclides in the approx. 500 ml of material sample provided by the organizer laboratory (ITM, Madrid, Spain), both in quantity and quality.

[3] SIRA: Samling and Identification of Radiological Agents

Sample was prepared by organized spiking the total volume of ultra pure water ("Milli-Q" quality) acidified with ultra pure HNO_3 (analysis quality as stabilizer) with aliquots of weighed standards. After homogenization sub-samples were taken to test the sample homogeneity by liquid scintillation counter (LSC). Samples (Table 2) were prepared with the aim to allow the laboratories measure in a similar geometry to 500 ml Marinelli in a plastic container.

In the postage box we received all the needed information to participate in the exercise (sample description, the exercise written instructions and scenario).

According to the exercise rules the primary results were to be sent to the organizer within 24 hours of receipt via e-mail, with confirming radio analytical results to follow within a week, also via e-mail for a fast information exchange. The Hungary was represented by the Hungarian Defence Forces Radiological Laboratory (HDF RL), with the Paks Nuclear Power Plant (PNPP) and the Agricultural Management Bureau Centre's Reference Laboratory for Radio analysis (AMBC RLR) acting as technical advisor and providing control measurements.

The organizing laboratory after the laboratory test reported the liquid sample content, which contained two radionuclides. The participating laboratories (with different anonym laboratory codes A-N) located the Am-241 isotope with gamma spectrometric measuring system with semiconducting detectors and the Pu-239 activity with alpha spectroscopy.

The reference activity of the sample was 3.94 ± 0.16 Bq/kg in case of the Am-241 radioisotope. In view of the 24 hour measurement results only 5 of 13 laboratories reached the given ± 10% (± 0.39 Bq/kg) criteria, while in view of the week long measurements this figure was 7 of 13 (Figure 1 and 2).

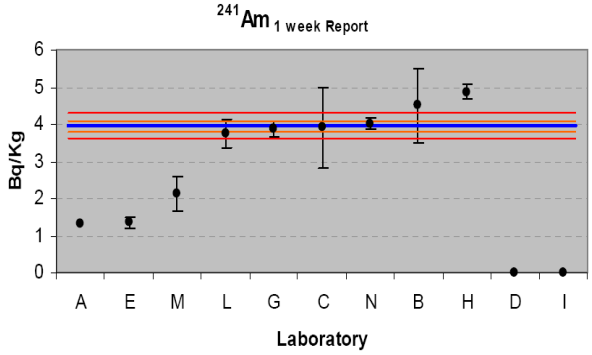

Figure 1.: Am-241 24 hrs. results (op. time)

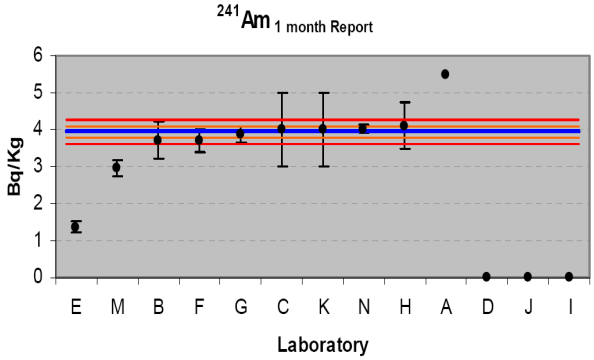

Figure 2. Am-241 1 week results (op. time)

Note: Hungarian Defence Forces Radiological Laboratory (HDF RL) code was K.

The activity of the Pu-239 (0.1% Pu-240 contamination) was 15.47 ± 0.72 Bq/kg. In 2 laboratories, the results of the 24 hour measurements were up to the stringent requirements of the ± 10% (± 1.55 Bq/kg), the number for the week long measurements was barely 3 (Figure 3 and 4).

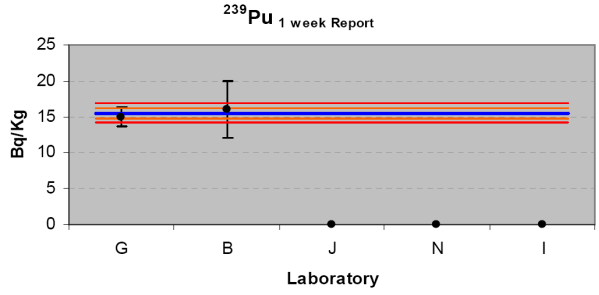

Figure 3. Pu-239 24 hrs. results (op. time)

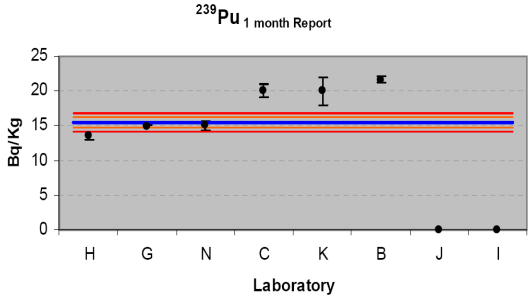

Figure 4. Pu-239 1 week results (op. time)

The identification and quantification of the Am-241 was made correctly by most of the participants. The identification of Pu-239 presented a higher difficulty for the laboratories, 3 laboratories had shown a perfect result in the identification and quantification.

In the Table 2 below we present the reported results by participating laboratories. The results have been normalized to Bq/kg units. Density of sample employed to normalize result from Bq/l to Bq/kg was 1.094 ± 0.001 kg/l.

Sample number/Lab. code	**24 h Report (Normalized Result)**		**1 Month Report (Normalized Result)**	
	Detected isotopes	**Certified Activity** [Bq/kg]	**Detected isotopes**	**Certified Activity** [Bq/kg]
08257/A	Am-241	1.3	Am-241	5.46
08257/B	Am-241 Pu-239	4.5±1.0 16±4	Am-241 Pu-239	3.7±0.5 21.6±0.5
08257/D	Am-241	Qualitative	Am-241	Qualitative
08257/E	Am-241	1.36±0.15	Am-241	1.36±0.15
08257/F	none		Am-241	3.7±0.3
08257/G	Am-241 Pu-239+Pu-240	3.88±0.21 15.0±1.3	Am-241 Pu-239+Pu-240	3.88±0.21 14.9±0.2
08257/H	Am-241	4.87±0.21	Am-241 Pu-239	4.1±0.62 13.5±0.54
08257/I	Pu-239 Am-241	Qualitative Qualitative	Pu-239 Am-241	Qualitative Qualitative
Sample number/Lab. code	**24 h Report (Normalized Result)**	**1 Month Report (Normalized Result)**	**Sample number/Lab. code**	**24 h Report (Normalized Result)**
08257/J	Pu-239	Qualitative	Pu-239 Am-241	Qualitative Qualitative
08257/K	none		Am-241 Pu-239	4±1 20±2
08257/L	Am-241	3.74±0.38	No participation	
08257/M	Am-241	2.13±0.45	Am-241	2.96±0.21
08257/N	Am-241 Pu-239+Pu-240	4.01±0.14 Qualitative	Am-241 Pu-239+Pu-240	4.01±0.11 15.0±0.6

Note: the Hungarian laboratory code (HDF RL) was K.

Table 2. Radioanalytical normalized results (cont.)

The results of this exercise according to Table 2 results shown a high heterogeneity between NATO radiological laboratories in resources, capabilities and working experience. In Table 3 we summarize the alpha and gamma spectrometry measuring conditions, method and equipment, which used the Hungarian laboratory.

Laboratory short name	Methods and equipments	
	Alpha spectrometry	**Gamma spectrometry**
HDF RL (K)	No equipment available*	Sample preparation: none Spectrometer: Canberra GC2020 coax. HpGe Geometry: 500 ml Marinelli, Pb shielding Measuring time: 24 h Analyzer: Inspector2000 Software: Genie2000 *Result:* Am-241 detected
AMBC RLR	Sample preparation: 10,00 cm^3 sample evaporation Spectrometer: Canberra Software: Canberra Apex Alpha Measuring time: 12 h Result: Am-241 and Pu-239	Sample preparation: none Spectrometer: Canberra HpGe Geometry: 500 ml Marinelli, Canberra Pb shielding Measuring time: 33,33 h Software: Winner 6.0 (FAST ComTec GmbH) *Result:* Am-241 detected
Laboratory short name	**Alpha spectrometry**	**Gamma spectrometry**
PNPP CES	Sample preparation: 4,00 cm^3 sample evaporation Spectrometer: Alpha Analyst M7200-04, 8 chambers Software: Canberra Apex Alpha Measuring time: 12 h Result: Am-241 and Pu-239	Sample preparation: none Spectrometer: Canberra HpGe Geometry: 10 ml plastic container Measuring time: 8,33 h Software: Canberra APEX Result: Below the detection limit

Table 3. Hungarian sample preparation, conditions and equipment (1 week) (cont.)

Abbreviations to Table 3:

HDF RL: Hungarian Defence Forces Radiological Laboratory

PNPP CES: Paks Nuclear Power Plant Operations Division, Chemical Engineering Section

AMBC RLR: Agricultural Management Bureau Center's Reference Laboratory for Radio analysis

In a Table 4 we present the three Hungarian laboratory control measurement results.

Nuclide	**ITM standard** [Bq/kg]	**HDF RL** [Bq/kg]	**AMBC RLR** [Bq/kg]	**PNPP CES** [Bq/kg]
Am-241	3,94 ± 0,16	4,0 ± 1	3,8 ± 0,2	3,66 ± 0,46
Pu-239 (Pu-239 + Pu-240)	15,47 ± 0,72	AMBC RLR result	20,1 ± 2,0	17,8 ± 1

Table 4. Hungarian radioanalytical results

*Note: HDF Radiological Laboratory doesn't have a liquid scintillation counter, for Pu-239 we presented the AMBC RLR results and figures.

Events show, that in spite of the different instruments and methods of measurement, within the results of the three Hungarian laboratories, with scatter adjustment, are whithin the reference values published by the organising laboratory.

The organizer did the evaluation of the laboratories' results according to the ISO 13528 (2005) and ISO/IEC 43 guide (1997) the accepted limits of the measurements were ±10 % of the actual value.

Main statistic proposed:

U-score/E_n numbers: $$E_n = \frac{x - X}{\sqrt{u_x^2 + U_x^2}}$$

Where:
x: reported value of participant laboratory,
X: assigned value determined in a reference laboratory,
U_x: expanded uncertainty of X,
u_x: expanded uncertainty of a participant's result.

u-score acceptance criteria: unsatisfactory $|u| > 2.58$
satisfactory $|u| < 2.58$

U-score								
Laboratory code	**24 hours (operational time)**				**1 week (operational time)**			
	Am-241		**Pu-239**		**Am-241**		**Pu-239**	
A(1)	Det.				Det.			
B(2)	0.19	**< 2.58**	0.23	**< 2.58**	0.46	**< 2.58**	7.00	**> 2.58**
C(3)	0.36	**< 2.58**			0.31	**< 2.58**	2.41	**< 2.58**
D(4)	Det.				Det.			
E(5)	12.7	**> 2.58**			12.7	**> 2.58**		
F(6)					1.75	**< 2.58**		
G(7)	0.23	**< 2.58**	0.32	**< 2.58**	0.23	**< 2.58**	0.78	**< 2.58**
H(8)	3.5	**> 2.58**			0.25	**< 2.58**	2.20	**< 2.58**
I(9)	Det.		Det.		Det.		Det.	
J(10)			Det.		Det.		Det.	
K(11)					0.06	**< 2.58**	2.13	**< 2.58**
L(12)	1.36	**< 2.58**						
M(13)	4.51	**> 2.58**			4.92	**> 2.58**		
N(14)	0.32	**< 2.58**	Det.		0.35	**< 2.58**	0.33	**< 2.58**

Table 5. U-score results

Legend and comments: "Det": "Detected" the participant has reported the isotope qualitative, but he has not given an estimation of uncertainty that allows U-score calculus. Light grey: no data, dark grey: outside the accepted value (unsatisfactory, $u > 2.58$), diagonal striped: Satisfactory measured result ($u < 2.58$)

The participation of this proficiency test exercise was very useful for all laboratories to test laboratory capabilities and compare the results. We planned to participate in the future on the next NATO SIRA exercise.

CONCLUSION

Most of the participating laboratories were successful in completing the measurement exercise and taking part in the SIRA-2008 exercise. Spectrometric identification of the Am-241 gamma did not pose much of a problem for most of the laboratories. Based on the 24 hour measurement results of the Pu-239 alpha spectrometric identification, altogether two laboratories were successful, based on the one week measurement results, three laboratories were fully within the acceptance criteria (measurement time could be increased). Knowing the low alpha activity concentration of the sample the primarily communicated results are acceptable, since the organizers gave extremely strict acceptability criteria.

The main reason for the failure was that the laboratories did not publish results of the measurements, did not publish standard deviation or that their results were outside the stringent ± 10% limit. In samples of small concentration of alpha radiating activity (below 20 Bq/kg) we consider a larger margin (± 25%) to be acceptable. Most of the participating laboratories were well equipped, but HDF RL not equipped with alpha spectrometer. In the future the main task is to develop technical and human resources in HDF RL according to SIRA Handbook (AEP-49) recommendations. [3]

ACKNOWLEDGEMENTS

We wish to acknowledge the result and the contributions of organizing and participating laboratories in the SIRA exercise: Raul Lopez S (ITM "La Maranosa", Spain), Elizabeth Inrig (Defence R&D, Canada), Petr Sladek (NBC Defence Institute, Czech Republic, N. Chianea (SPRA, France), M. Kitto (Deployable NBC Analytical Laboratory, Germany), A. MAssaro ()entro Tecnico Logistico Interforze NBC, Italy), Gary Mallard (OPCW Laboratory, The Netherlands), B. Hanne (Norwegian Defence Research Establishment FFI, Norway), M. Ceremuga (Military Institute of Chemistry and Radiometry, Poland), K. Lizon (RCBO Reference Chemical Laboratory, Slovakia), T. Annika (FOI, Sweden), M. Simpson (DSTL, United Kingdom), T. Paul (AWE, United Kingdom), S. Tarján (AMBC RLR, Hungary),

REFERENCES

[1] Haslip, Dean S.; Mercier, J. R.: A NATO Exercise on Radiological Sampling, Health Physics, November 2004, Volume 87, pp. S63-S67

[2] NATO AEP-49 Sampling and Identification of Radiological Agents (SIRA) Handbook, 2004, (NATO/PfP Unclassified)

[3] Raul Lopez S.: 2008 NATO SIRA Laboratory Exercise Technical Report, La Maranosa, Spain, 2009 (NATO/PfP Unclassified)