IAEA 60 Years
Atoms for Peace and Development

IAEA's ALMERA Effort towards Harmonization of Radioanalytical Procedures

Development and Validation of a Rapid Procedure for Simultaneous Determination of ^{89}Sr and ^{90}Sr in Soil Samples using Cerenkov and LSC

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International Atomic Energy Agency

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Outline

- **Overview of the analytical method development activity in the context of the IAEA's ALMERA network**
- **ALMERA developed and validated analytical methods for routine environmental monitoring and emergency situations**
- **Approach, development and validation of the rapid method for simultaneous determination of ^{89}Sr and ^{90}Sr in soil samples using Cerenkov and LSC**
- **Requirements and effort for harmonization of radioanalytical procedures**

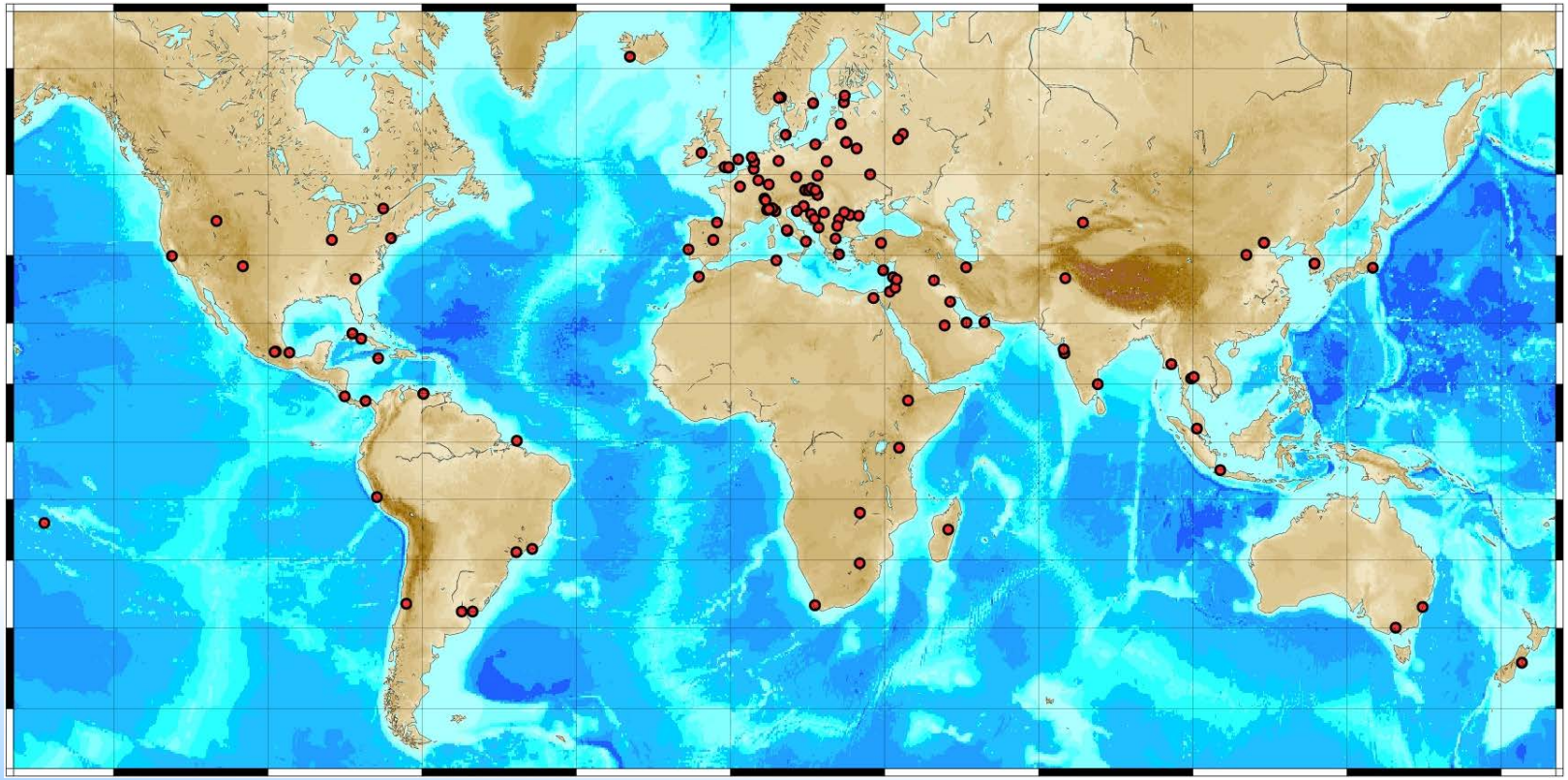
IAEA's ALMERA network

- **ALMERA:** IAEA's Network of Analytical Laboratories for the Measurement of Environmental Radioactivity
- A worldwide network of laboratories capable of providing reliable and timely analysis of environmental samples in the event of an accidental or intentional release of radioactivity in the environment

Development of ALMERA

- **1995:** Establishment of ALMERA
- **1997:** 1st ALMERA coordination meeting
- **1999:** First official nominations of laboratories

April 2017: 160 laboratories from 87 Member States



ALMERA developed and validated analytical methods

Sample matrix	Radionuclides							
	Pu isot.	^{241}Am	^{90}Sr	^{210}Po	^{210}Pb	$^{226+228}$Ra	U	Th
Aerosol	O	O	O					
Soil/sediment	O/O	O/O	O/O					
Terrestrial surface water				O		O		
Milk			O					
Phosphogypsum				O	O	O	O	O

O: Routine procedure for routine environmental monitoring

O: Rapid method for emergency situations

All IAEA developed and validated analytical methods available as pdf files under:

https://nucleus.iaea.org/rpst/ReferenceProducts/ALMERA/Validated_analytical_methods/index.htm

Routine methods for environmental monitoring

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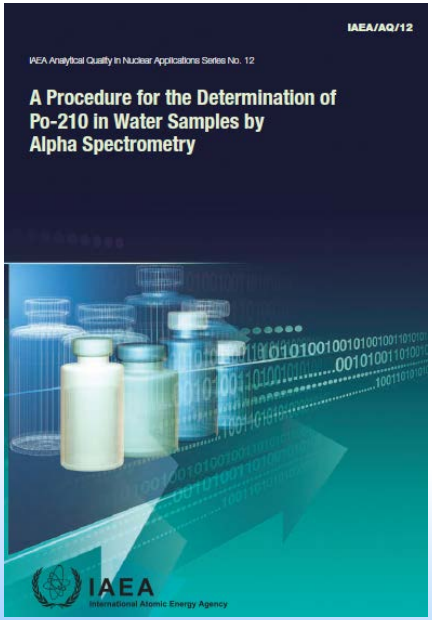
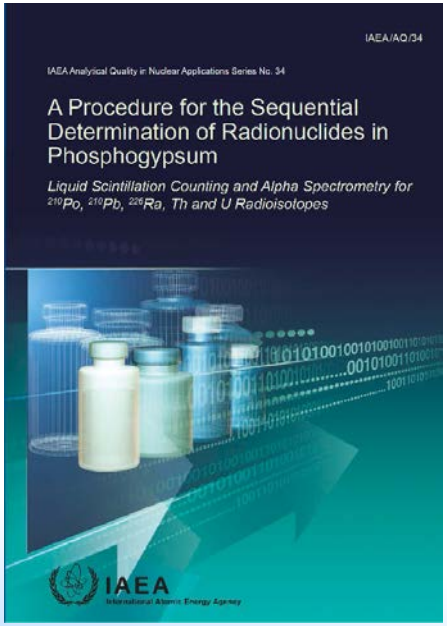
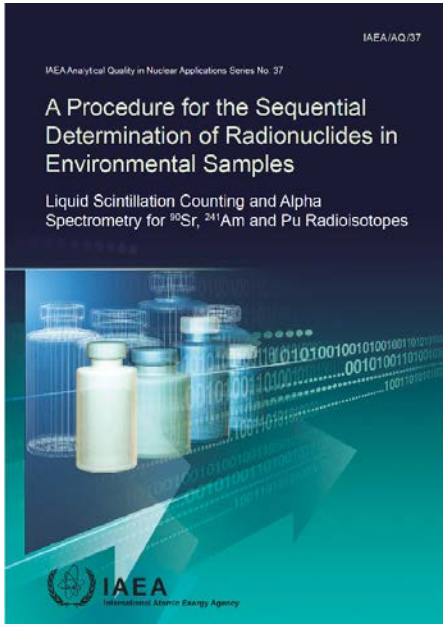
Three methods for routine environmental monitoring developed and validated within the ALMERA network :

- **Determination of ^{210}Po in water samples;**
- **Determination of ^{210}Po, ^{210}Pb, ^{226}Ra, Th and U radioisotopes in phosphogypsum;**
- **Determination of ^{90}Sr, ^{241}Am, and Pu radioisotopes in environmental samples.**



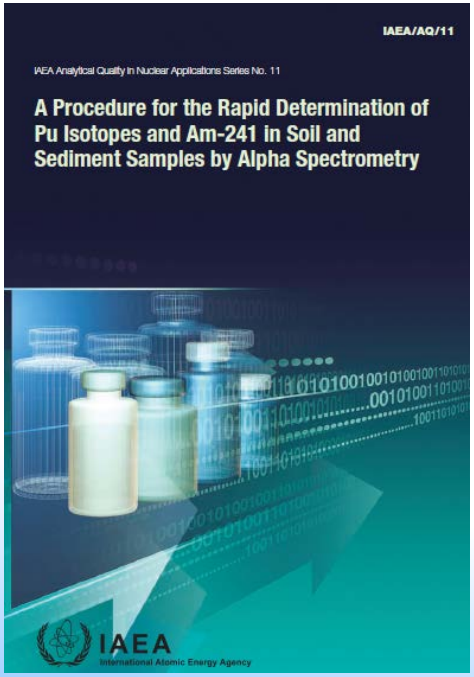


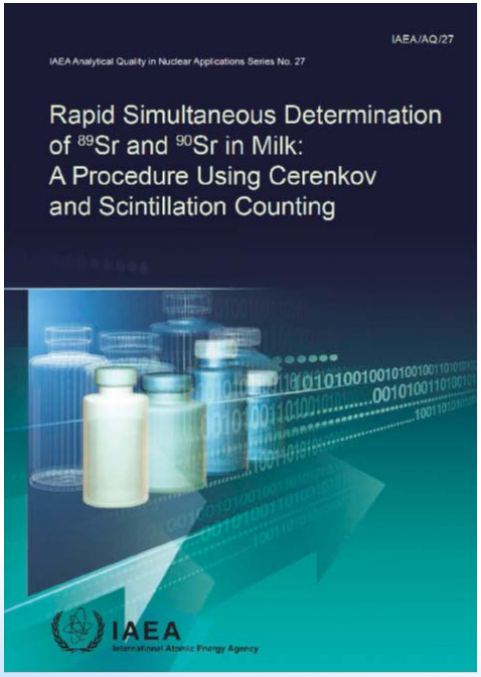


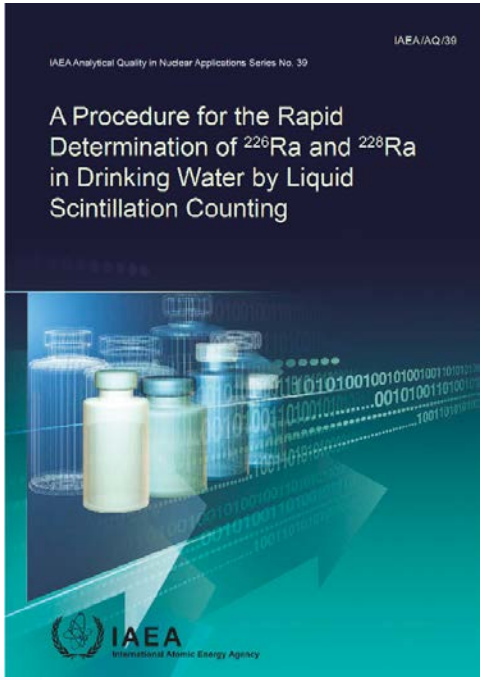
Rapid methods for emergency situations

Three rapid methods for emergency monitoring developed and validated within the ALMERA network:

- **Rapid determination of Pu Radioisotopes and ^{241}Am in soil and sediment samples;**
- **Rapid determination of ^{89}Sr and ^{90}Sr in milk samples;**
- **Rapid determination of ^{226}Ra and ^{228}Ra in drinking water.**







‘Hands-on’ training courses on methods

Checking for Radionuclides in Dairy Food Products

ALMERA Training Course Demonstrates State-of-the-art Method for Detecting Radiostrontium in Milk

By Aurelien Pitois, , IAEA Environment Laboratories

07
November 2014

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Experts from the IAEA and Korea Institute of Nuclear Safety train scientists on a rapid analytical method to be used for food monitoring in emergency situations at a training course held in Daejon from 3 to 7 November 2014. (Photo Credit: A. Pitois/IAEA)

ALMERA ‘hands-on’ training courses on published ALMERA rapid methods in radiochemical laboratories

➡ **Approx. 60 ALMERA staff members trained in the last 3 years**

IAEA and Argonne National Laboratory Train Scientists on State-of-the-Art Methods for Rapid Environmental Radioactivity Assessment

For two weeks in March, 24 scientists representing laboratories in 21 countries learned state-of-the-art methods for precise and quick assessment of radionuclides in the environment. The venue was a training course on rapid assessment methods for environmental radioactivity, held between 10 and 21 March 2014 at the Argonne National Laboratory (ANL) in the US. It was organized by the Environment Laboratories Division in the IAEA Department of Nuclear Sciences and Applications in cooperation with the ANL to particularly benefit scientists working in laboratories belonging to the ALMERA network.

ALMERA is the acronym for Analytical Laboratories for the Measurement of Environmental Radioactivity, a world-wide network of analytical laboratories capable of providing reliable and timely determination of radionuclides in

Rapid method for ^{89}Sr and ^{90}Sr in soil

Why the rapid determination of ^{89}Sr and ^{90}Sr in soil?

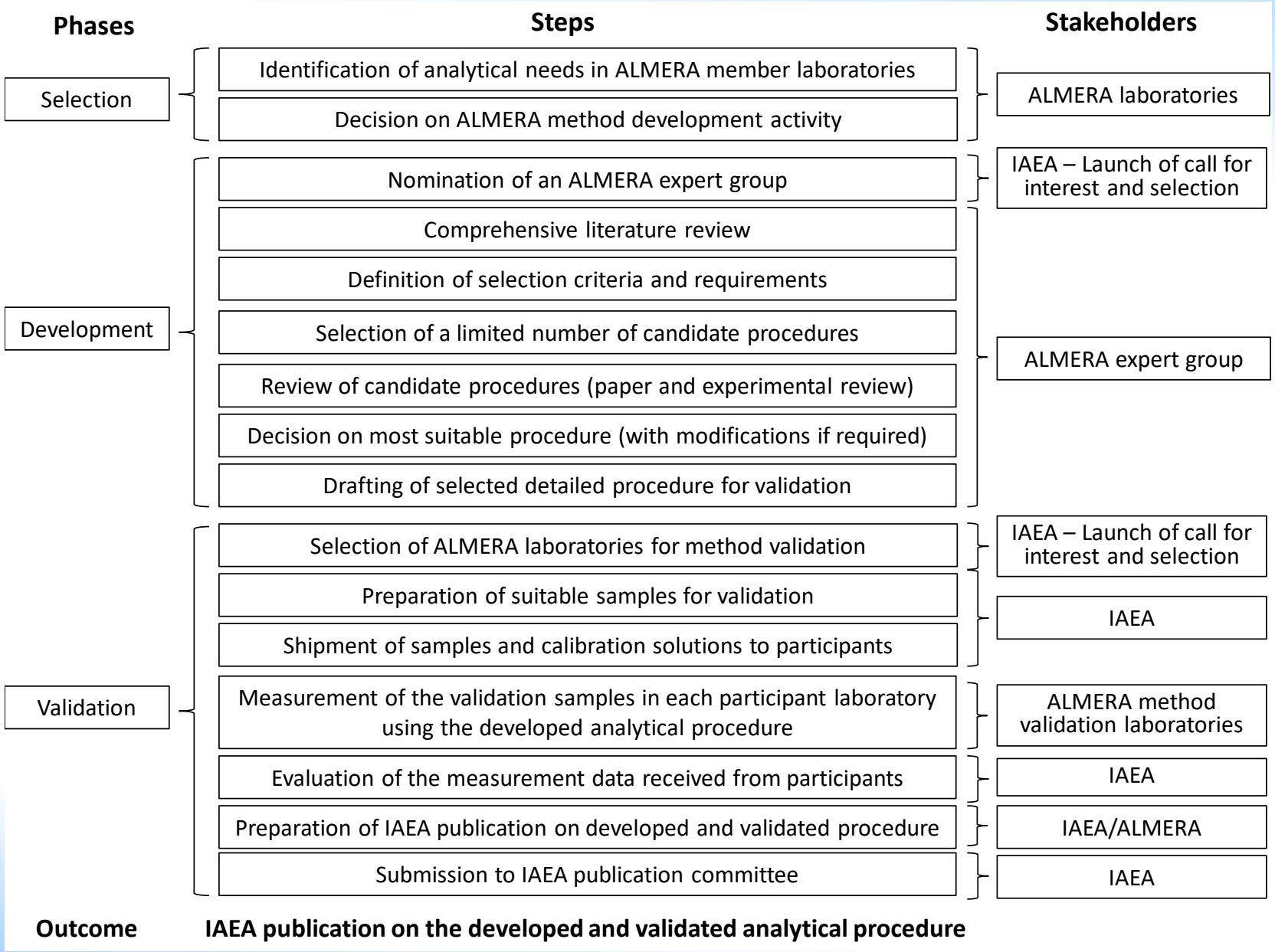
- **^{89}Sr (half-life 51 days) and ^{90}Sr need to be measured immediately after a nuclear or radiological accident, since the dose is mainly coming from ^{89}Sr in the first weeks**

  ➡ **Important for emergency response situations**

- **Radiostrontium is one of the most biologically hazardous radionuclides, since it becomes incorporated in the calcium pool.**

- **Proficiency test results show analytical difficulties for laboratories to measure ^{90}Sr in soil.**

Approach used



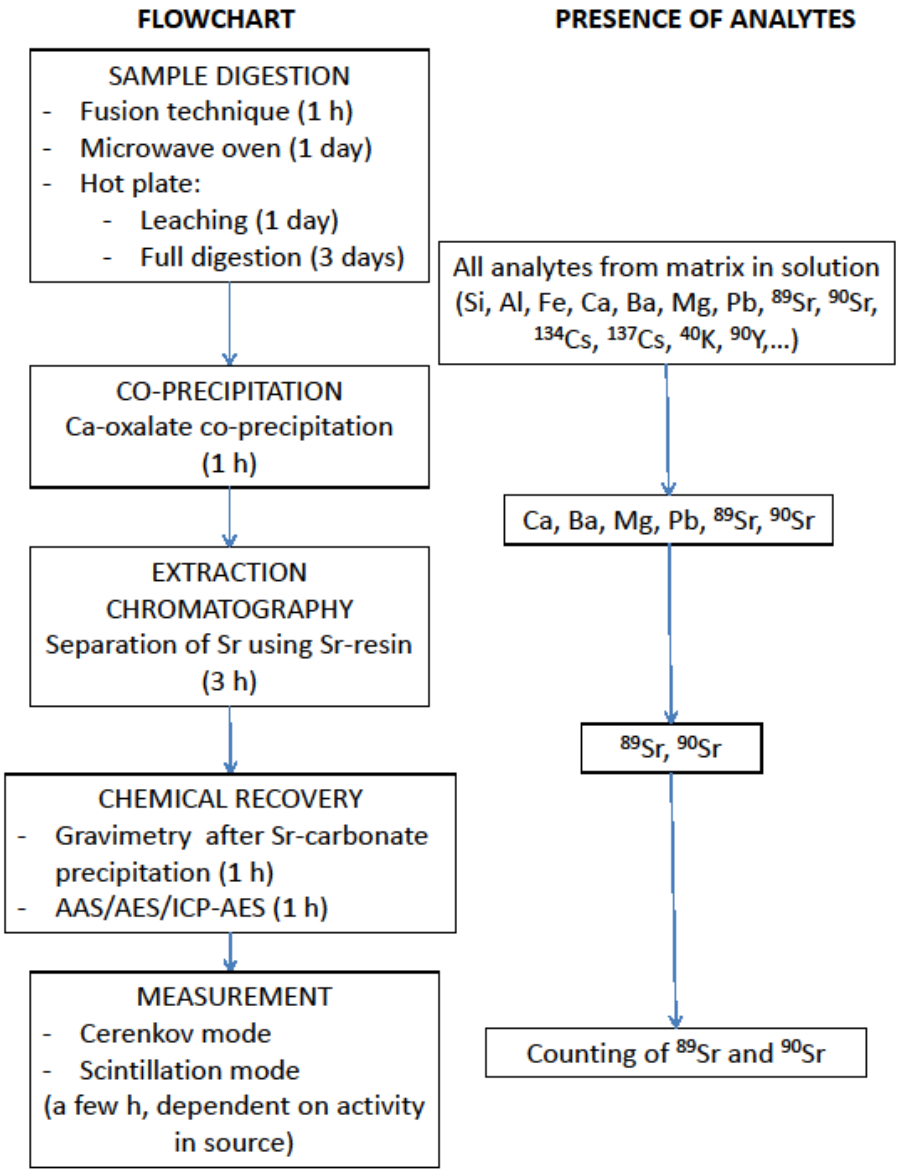
Development phase

- **ALMERA expert group:**
  - **ANSTO, Australia;**
  - **University of Basque Country, Spain;**
  - **Spiez Laboratory, Switzerland;**
  - **Korea Institute of Nuclear Safety, Republic of Korea (IAEA Collaborating Centre);**
  - **IAEA, Seibersdorf (Coordinator).**
- **Comprehensive bibliography on the determination of radiostrontium in soil**
- **Definition of selection criteria and requirements**

Development phase

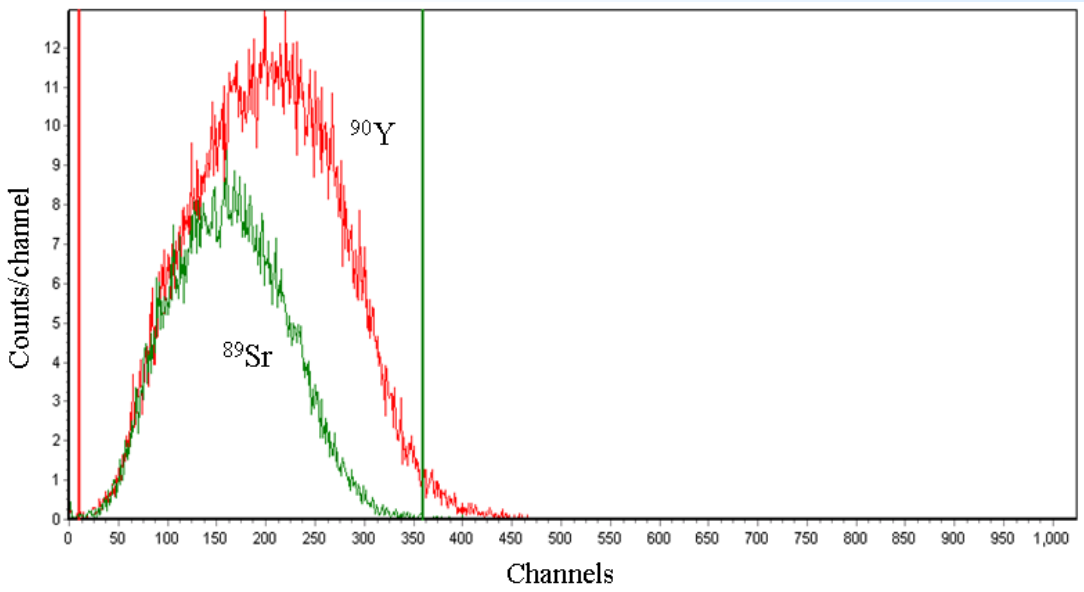
- **Primary selection criteria:**
  - **Useful in emergency situations ($^{89}Sr/^{90}Sr$);**
  - **Rapidity of the method;**
  - **Efficiency of the chemical separation;**
  - **Potential use in a wide range of laboratories.**
- **Secondary selection criteria:**
  - **Detection limit;**
  - **Number of samples to be handled in parallel;**
  - **Cost effective;**
  - **No use of fuming nitric acid.**

Development phase
Flow chart of the procedure

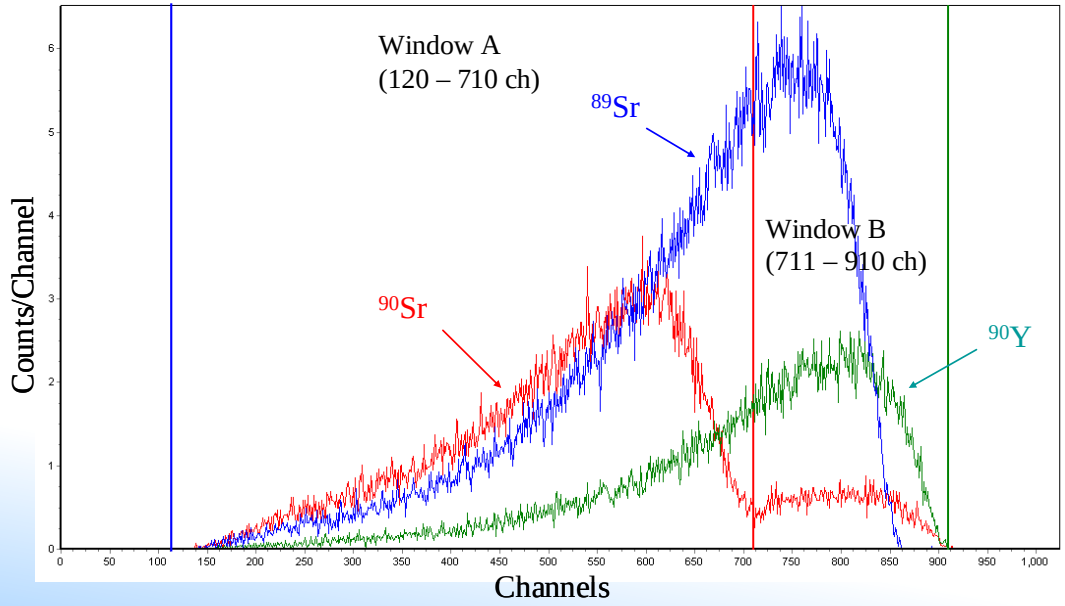


Drafting of detailed procedure for validation, after comprehensive paper evaluation and experimental review.

Typical spectra from measurement process



Measurement in Cerenkov mode



Measurement in Liquid Scintillation Counting mode

Validation phase

- **Interest expressed by 23 laboratories from 20 countries**
- **Selection of 10 laboratories:**
  - **Laboratories from the expert group (ANSTO, Uni. Basque Country, Spiez Laboratory, KINS, IAEA Seibersdorf)**
  - **Federal Office for Radiation Protection (BfS), Germany;**
  - **Bhabha Atomic Research Centre (BARC), India**
  - **Japan Chemical Analysis Center (JCAC), Japan**
  - **Korea Atomic Energy Research Centre (KAERI), Republic of Korea;**
  - **Swedish Defence Research Agency (FOI), Sweden;**
  - **Turkish Atomic Energy Authority (TAEK), Turkey.**

Validation phase

- **5 validation samples prepared at IAEA, Seibersdorf + 2 ampoules for LSC calibration + blank**
  - **Soil spiked with $^{89}Sr/^{90}Sr$~ 5 (to check robustness of the method);**
  - **Soil spiked with $^{89}Sr/^{90}Sr$ ~ 10 (to check robustness of the method);**
  - **Soil spiked with $^{89}Sr/^{90}Sr$ ~ 20 (to define limitations of the method at high ratio);**
  - **Soil spiked with $^{89}Sr/^{90}Sr$ ~ 5 at low massic activities (to check sensitivity of the rapid method);**
  - **Soil spiked with $^{89}Sr/^{90}Sr$ ~ 5 with heat treatment (potentially embedded radiostrontium) (to define limitations of the method in terms of digestion methods);**
  - **1 ampoule of ^{89}Sr for calibration of the LSC;**
  - **1 ampoule of ^{90}Sr for calibration of the LSC;**
  - **1 unspiked soil (blank)**

Validation phase

- **Shipment of validation samples to participants;**
- **Measurement of the validation samples in the laboratories using chosen digestion techniques:**
  - **Fusion technique: 7 laboratories;**
  - **Microwave oven digestion: 6 laboratories;**
  - **Leaching using hot plates: 6 laboratories;**
  - **Full digestion using hot plates: 4 laboratories.**

In total, 230 measurement data!

Comprehensive validation campaign
to ensure robustness and confidence

Some validation results: outlier tests

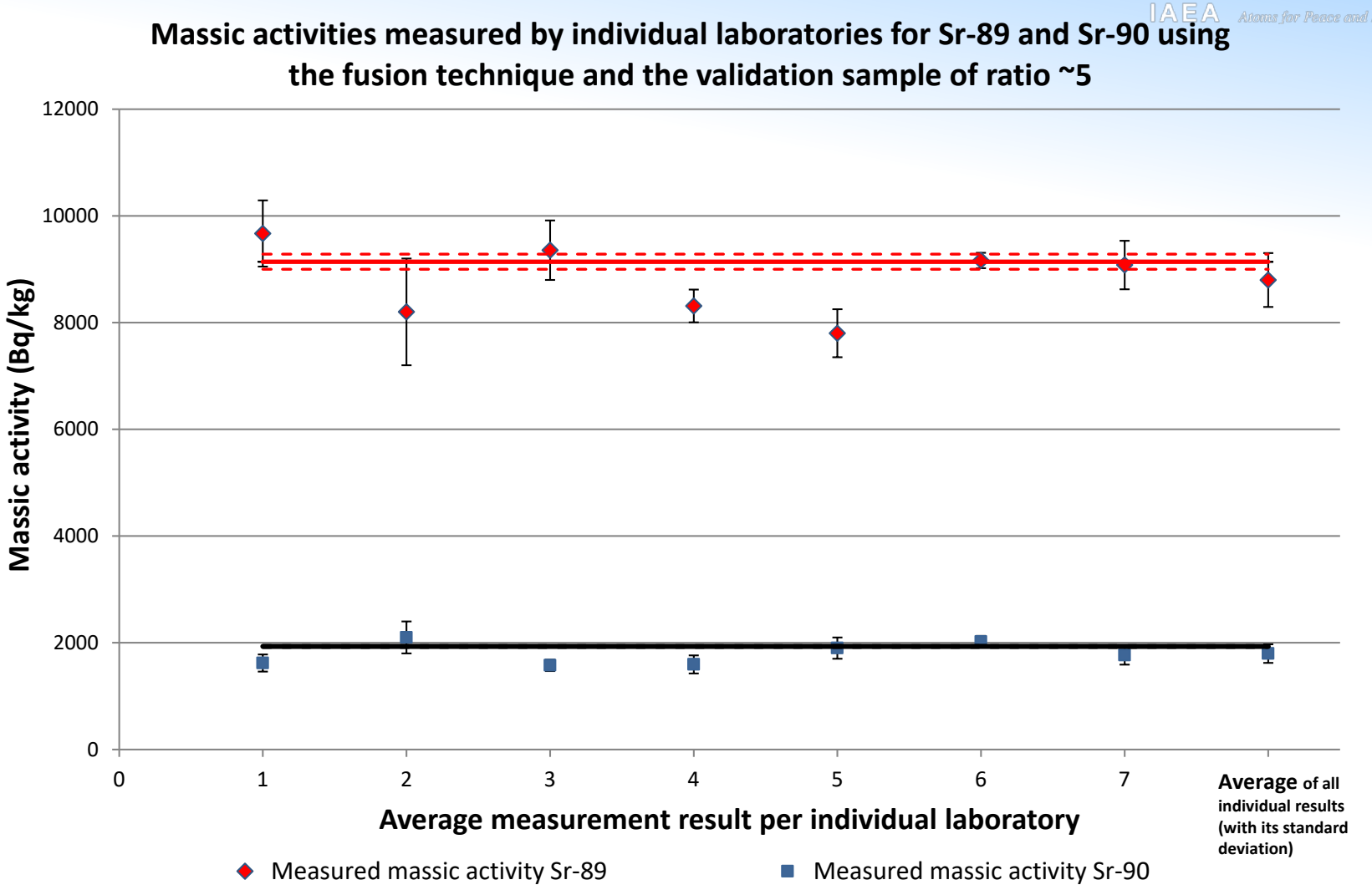
- **Several statistical tests on distribution:**
  - **Dixon;**
  - **Grubbs;**
  - **Skewness;**
  - **Kurtosis.**

- **Test on relative bias on target values:**
  - **Setup value: >45% for ratio 5 and 10;**
  - **Setup value: >65% for ratio 20.**

→ **18 outliers:**
  - **12 from statistical tests;**
  - **6 from test on relative bias.**

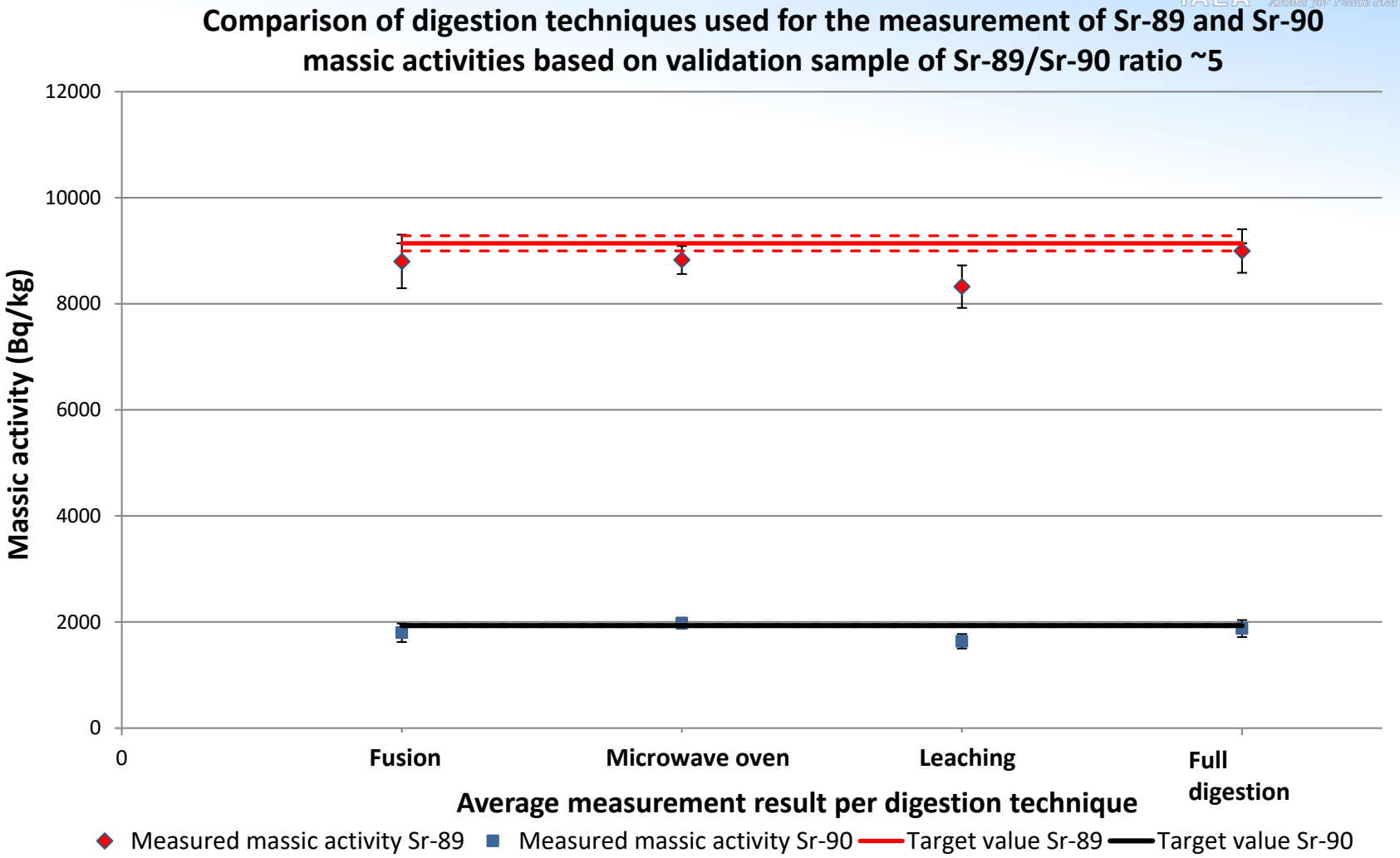
→ **7% outliers on the total number of measurement data (removed from further data analysis)**

Some validation results: individual results



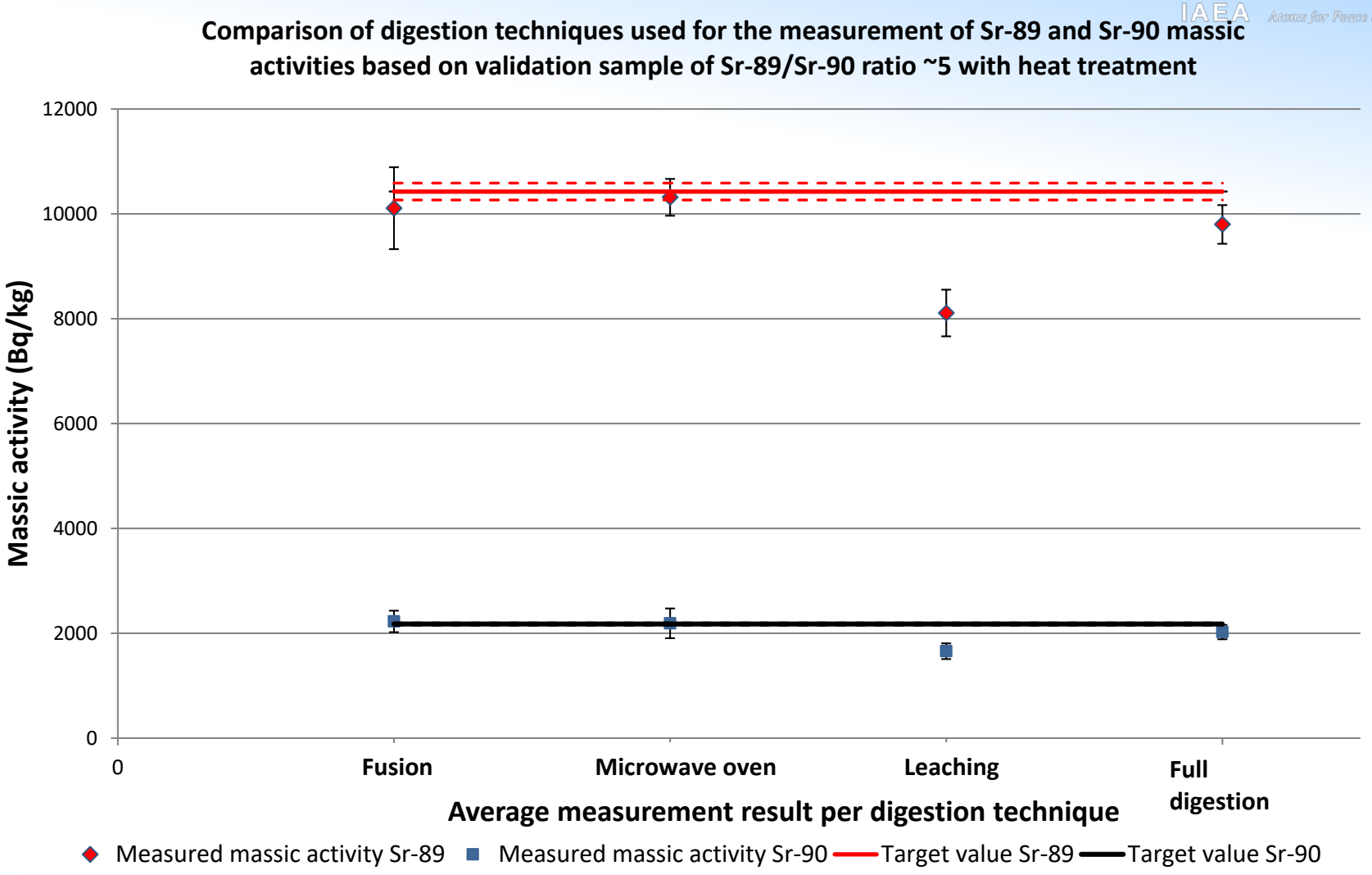
- Good agreement of the individually reported measurement results with the target value
- Average of all individual results given in last position with its standard deviation and used for comparing the results obtained by the different digestion techniques (see next slide).

Comparison of digestion techniques



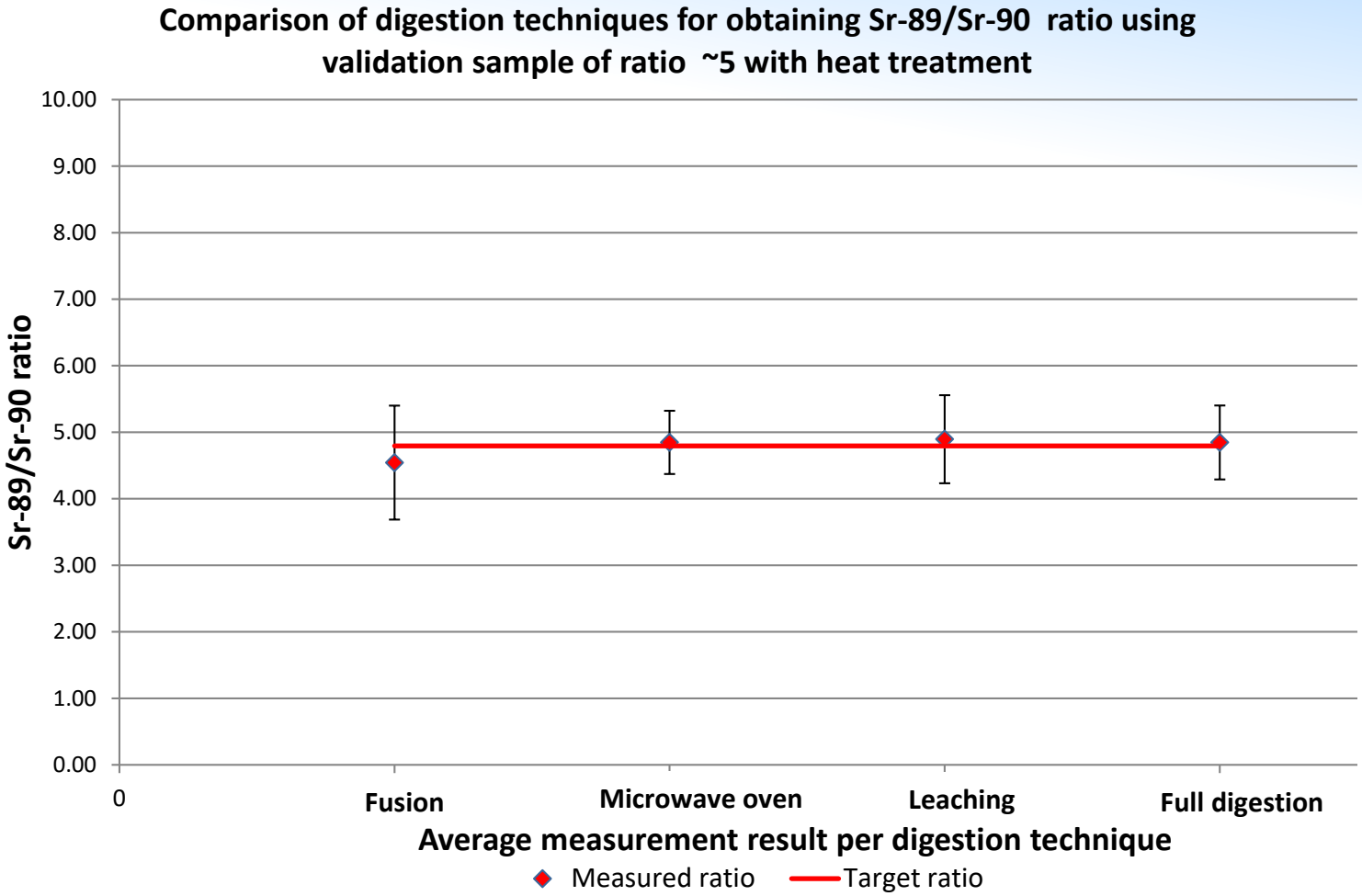
- Good agreement for all used digestion techniques, except leaching
- Underestimation of ~10% using the leaching technique (observed for all ratios)
- Could still be acceptable for emergency response however.

Comparison of digestion techniques



- Heat treatment: Attempt to simulate potentially embedded radiostrontium
- Good agreement for all used digestion techniques, except leaching;
- Underestimation of ~25% using the leaching technique, **not suitable digestion technique (incomplete dissolution)**

Measured $^{89}Sr/^{90}Sr$ ratios



- Good agreement between measured and target $^{89}Sr/^{90}Sr$ ratios for all digestion techniques
- True too for validation sample with heat treatment

Conclusions from the validation phase

- **Robust method? Yes,** overall good agreement of the individually reported measurement results with the target values (checked on 230 measurement data), $^{89}Sr/^{90}Sr$ ratio well estimated

- **Method suitable at high $^{89}Sr/^{90}Sr$ ratios: Yes,** good agreement between measured and target values for all investigated ratios, i.e. 5,10 and 20

- **Sensitivity?** Method checked at lower activity range with good agreement between measured and target values

- **Limitations in terms of digestion techniques?**
  - **Fusion, microwave oven, full digestion:** good agreement between measured and target values/ratios for all validation samples
  - **Leaching:**
    - Underestimation of ~10% in the absence of embedded radiostrontium, could still be acceptable in emergency response
    - Underestimation of ~25% (or more) in the presence of embedded radiostrontium/refractory material, not suitable digestion technique
    - Measured and target $^{89}Sr/^{90}Sr$ ratios are in good agreement for all validation sample (in agreement with incomplete dissolution, same behaviour for ^{89}Sr and ^{90}Sr)

Conclusions

- **Development and validation of a rapid procedure for the simultaneous determination of ^{89}Sr and ^{90}Sr in soil samples using Cerenkov and LSC counting ⇨ Publication under drafting**

- **Requirements for harmonization of radioanalytical procedures:**
  - **Methods recognized and accepted by the scientific community;**
  - **Comprehensive and clearly formulated methods;**
  - **Large availability to all interested parties;**
  - **Easy application in a large number of laboratories worldwide.**

- **ALMERA effort for harmonization of radioanalytical procedures:**
  - **Strong involvement of laboratories worldwide with recognized expertise and geographical balance: ALMERA member laboratories;**
  - **Use of a thorough methodology;**
  - **Publications free of charge and available for download;**
  - **'Hands-on' training courses for ALMERA members.**

- **A big thank you to all ALMERA contributors!!!**

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