ISSN 1831-9424

European Commission

European survey of measuring natural radioactivity in building materials

Feedback from the REM 2020 proficiency test

Hult, M., Jobbágy, V.

2025

Joint Research Centre

EUR 40277

Contact information
Name: Mikael Hult
Address: EC-JRC, Retieseweg 111, 2440 Geel
Email: Mikael.hult@ec.europa.eu
Tel.: +32-14-571269

EU Science Hub
https://joint-research-centre.ec.europa.eu

JRC141062

EUR 40277

PDF ISBN 978-92-68-26021-0 ISSN 1831-9424 doi:10.2760/7252222 KJ-01-25-205-EN-N

Luxembourg: Publications Office of the European Union, 2025

How to cite this report: European Commission: Joint Research Centre, Hult, M. and Jobbágy, V., European survey of measuring natural radioactivity in building materials, Publications Office of the European Union, Luxembourg, 2025, https://data.europa.eu/doi/10.2760/7252222, JRC141062.

Contents

Abstract

The Joint Research Centre of the European Commission organised a proficiency test (PT) on the measurements of natural radioactivity (U-238, Ra-226, Pb-210, Th-232, Ra-228, Th-228 and K-40) in two building materials. The 105 registered laboratories were requested to submit measurement results using their routine analytical procedure. In addition, participants were requested to provide information about their organisation, routine applied procedure and future PT needs via on-line questionnaires. For this purpose, two on-line questionnaires were set up. The first questionnaire was sent during registration (registration-survey) and the other one (reporting-survey) was connected to submission of measurement results for the PT. Out of 105 laboratories, 96 submitted at least one result and 85 submitted the detailed reporting-survey. These questionnaires provide valuable information that can be further used to identify pitfalls, best practices and to improve analytical procedures in radiological laboratories. In addition, participants had the opportunity to give feedback on the PT and express their wishes which radionuclides and matrices they would consider useful in the future PTs. The most requested matrices were NORMs (Naturally Occurring Radioactive Materials) and water followed by air-filter and soil. In these matrices natural radionuclides are the most wanted ones, but there is a clear interest in artificial/transuranic radionuclides too.

Acknowledgements

Thanks are due all participating laboratories, as they invested their valuable resources and time to the REM 2020 PT participation and contribution to two on-line questionnaires with essential technical and organisational related information.

The initial coordinator for this PT was Katarzyna Sobiech-Matura, who also initiated the survey. Contributions to establishing the questionnaires were also given by Petya Malo, Raf Van Ammel and Jan Paepen at the Joint Research Centre, Nuclear Data and Measurement Standards Unit: (Unit G.II.6).

Author(s)

Mikael Hult - European Commission, JRC-Geel, Retieseweg 111, 2440 Geel, Belgium

Viktor Jobbágy - Social Organisation for Radioecological Cleanliness (SORC), Egyetem str 10, Veszprem 8200, Hungary

Executive summary

Policy context and concept

The European Commission's Joint Research Centre in Geel (JRC-Geel) on request of the Commission Directorate General for Energy (DG ENER) organised a proficiency test (PT) on the measurements of natural radioactivity in building materials (cement and expanded clay blocks). The JRC PTs are an integral part of the Commission's work of realising verifications of Member States' obligations linked to Article 35 of the Euratom Treaty. JRC-Geel organises PTs to assess the quality of the radioactivity measurements at Member State laboratories. The performance of methods and their scores in the building materials PT were already evaluated and published in a recent JRC report (Hult et al., 2024). Additionally, participants filled in two on-line questionnaires providing detailed information about their organisation and measurement methods. This report deals with analysis of the responses from the two questionnaires. The main aims of the analysis are to identify best practices, possible pitfalls and future needs in environmental radioactivity measurements to improve their quality.

Results

In this PT, 105 laboratories registered of which 96 laboratories submitted results and 85 filled in the on-line questionnaires. JRC-Geel received altogether 1586 measurement results (for seven radionuclides in three materials) and more than 18000 entries of additional data via questionnaires.

As seen in the questionnaires, approximately 60% of the participants measure naturally occurring radionuclides in building materials having 0 to 60 years of experience (average: 20 years). Among the participants, 22 are having legal obligations to perform these types of measurements. The annual number of samples varied between 1 and 400 (median: 20 samples) and all 85 participants followed their standard procedure during measurement of PT materials. Routinely about half of the laboratories determine moisture content in samples but in the REM2020 PT, 81 out of 85 (about 95%) participants measured it.

On one hand, around 95% of the measurement results yielded acceptable scores for precision. On the other hand, about one-third of the results did not have acceptable uncertainties and dubious detection limits were also indicated. The median detection limit values were <10 Bq/kg for all seven radionuclides. However, for Ra- 226 and K-40 detection limits of 15 - 1000 Bq/kg and 50 – 5000 Bq/kg were reported, respectively.

From the overall data, the relative expanded uncertainty values varied in the range of 0.5% - 96.6%. The median of the uncertainties for the radionuclides in ascending order is the following:
K-40 < Ra-228 < Ra-226 < Th-232 < Th-228 < U-238 < Pb-210.

When stricter assessment criteria (σ_{PT}) were used (15% instead or 20% for K- 40, Ra-226, Ra-228, Th- 228 and Th-232; 20% instead of 30% for U- 238 and Pb- 210) the number of satisfactory z scores did not change much, just decreased few percentage points, from 94.7% to 90.2%.

Main findings and conclusions

Member State laboratories generally measure building materials predominantly with non-destructive gamma-ray spectrometry method but some laboratories apply destructive analytical methods (alpha-particle spectrometry or ICP-MS/OES).

Samples are preferred to be received in form of powder or in small particles because less or no preparations are needed before measurements. When received as blocks (NORM-02), then sample preparation was tedious making difficulties to laboratories (dust formation, need for special tools).

Test portions were placed in well-defined geometries, radioactive equilibrium was considered by the participants. However, radon-tightness of the measurement beakers was tested only by 25 laboratories. Efficiency calibration was performed using Monte Carlo calculations, reference materials, mixed sources and also radioactive tracers. Participants indicated that further information on the material (density, chemical composition) would be also necessary to improve method accuracy and reduce uncertainties.

Based on the survey, a best sample preparation practice for gamma-ray spectrometry could be proposed. A general conclusion is that no major problems were identified for gamma-ray spectrometry and alpha-particle spectrometry. Also, solid-state scintillation and ICP-MS technique performed reliably in general. On the other hand, LSC, ICP-OES measurement methods were seriously underperforming.

Measurement uncertainties were under- as well as overestimated by participants. It was also found that U-238, Pb-210 and Th-228 measurements had the highest measurement uncertainties. In certain cases, unrealistic detection limits were also provided but that did not influence performance evaluation scores.

Gamma-ray spectrometry and alpha-particle spectrometry measurement methods were proved to be robust techniques with sufficient reproducibility and repeatability but alpha spectrometry exhibited better reproducibility for Th-228, Th-232 and U-238 measurements in all three PT materials.

After performing a simulation, it was found that stricter performance assessment criteria could be used in future for this type of PTs. As this was the first REM PT on building materials a conservative approach for σ_{PT} was taken since very little old PT-data was available. Therefore, another impact of this PT is that it can serve as an important benchmark for future PTs (not only with JRC as organiser) on radioactivity in building materials.

Related and future JRC work

Participants expressed their need for SI-traceable[1] radioactive reference materials and wide-scale proficiency tests in the environmental radioactivity field. JRC, as an impartial expert organisation, would play a vital role as PT organiser, reference materials provider and reference laboratory in this type of projects. Furthermore, topical workshops or training programs would be also beneficial for the participating laboratories and other stakeholders, emphasising good practices, uncertainty budgets and calculation of detection limits.

[1] Traceability of the measurement standards or measuring instruments to the International System of Units (SI) is established by means of an unbroken chain of calibrations or comparisons linking them to relevant primary standards of the SI units of measurements, Bq and kg in this case..

Quick guide

The JRC organised a proficiency test (PT) on building materials in support of Articles 35 and 39 of the Euratom Treaty. The 105 registered Member State laboratories were requested to report the measurement results of seven naturally occurring radionuclides in three materials (cement, expanded clay blocks and pulverised expanded clay blocks). Participants also submitted questionnaires including information about their organisation, procedures, needs on reference materials and future PTs; this way more than 18000 of additional data-entries was received.

This PT gives a representative snapshot primarily about the status of gamma-ray spectrometry measurements in this field as 91% of the submitted results were from gamma-ray spectrometry. Laboratories had difficulties to process bulky-hard material (NORM-02 block). Almost 95% of the total measurement results were acceptable in terms of precision. However, laboratories should review their uncertainty budget as in certain cases they under- or overestimated it. Some of the detection limits were unrealistically high, those participants should investigate whether the problem was with the applied method or made a mistake with detection limit calculation. It was also tested how the scores would change when stricter assessment criteria were used, still around 90% of the global measurement results would be acceptable. Therefore, these stricter criteria could be used in future for PTs on radioactivity in building materials.

1 Introduction

The REM[2] 2020 proficiency test on naturally occurring radionuclides in building materials was organised for the European radioactivity monitoring laboratories by the Joint Research Centre-Geel. This was the first of its kind PT focusing on building materials where participants were requested to measure naturally occurring radionuclides (U-238, Ra-226, Pb-210, Th-232, Ra-228, Th-228 and K-40) in two different building materials.

From the two building materials, three test items were produced in this PT; cement (named NORM- 01), pieces of expanded clay blocks (NORM-02) and the same expanded clay blocks but in pulverised form (NORM-03). The detailed proficiency test evaluation report including technical- and organisational details of the proficiency test was published in 2024 (Hult et al., 2024). The REM2020 PT technical report covered the PT test reference materials processing and production, assignment of reference values, the treatment of the reported data and provides details regarding the procedures used for the evaluation and comparison of the individual results with the assigned values. Furthermore, the participants could find the performance scores of every single submitted measurement results in that report which is important for some organisations from quality assurance and accreditation point of view.

It has to be noted that certain elements of this REM2020 PT exercise overlapped with the NORMCONSTRUCT project (Paepen et al., 2023a and 2023b) as the three test items NORM-01, NORM-02 and NORM-03 were used in both projects but for different purposes. Some standardised good practices for gamma-ray spectrometry measurements of construction products can be found in the afore-cited NORMCONSTRUCT reports by Paepen et al. (2023a and 2023b).

From 28 countries 105 laboratories registered to the REM2020 PT. However, not all could submit measurement results, the organizer received in total 1586 measurement results from 96 laboratories.

On top of the measurement results, the participants were requested to fill in two on-line questionnaires on a web application called EU Survey, one of the questionnaires was sent out in the time of registration and the other questionnaire was attached to the reporting. In these questionnaires the PT coordinator requested information on the participant laboratories' (i) experience, (ii) technical details on their methods and (iii) legal basis/obligation to perform these measurements. The registration survey collected information about the organisational settings, experience with NORM and construction materials measurements and general technical details about their routine measurement procedures. While the reporting survey focused more on technical details about the applied methods in this PT, for example sample preparation, calibration, detection limits, uncertainty budget and related difficulties. They could also indicate whether they made changes in their routine analytical procedures.

The registration and the reporting questionnaires were composed of four and five major sections respectively. The structure and the requested information of both surveys were similar but there were some differences as indicated in the list below. The titles of section 2 were different but in principle the targeted information was the same.

[2] REM stands for Radioactivity Environmental Monitoring and is the JRC programme on support to Article 35 and 39 of the Euratom Treaty

1. Contact details and type of the laboratory
2. Samples of building materials containing naturally occurring radionuclides(registration)/ General information on the performed measurements (reporting):
    - type and number of building material samples measured annually (registration)
    - year of experience (registration),
    - What kind of processing procedure and tools are applied in sample preparation
    - Moisture content determination
    - legal basis/obligation to perform these measurements (registration)
    - modifications comparing to the standard procedures (reporting)
3. Measurements methods:
    - Information about the naturally occurring radionuclides that are routinely measured
    - Applied measurement methods per nuclides
    - Sample and test portion preparation
    - Radon tightness
    - Efficiency calibration
    - Detection limits and calculations (reporting)
    - Uncertainty budget (reporting)
4. Problems encountered: whether the laboratories have any difficulties while measuring the naturally occurring radionuclides in building materials
5. Comments (reporting questionnaire)
    - Feedback on the REM2020 PT
    - Which radionuclides and matrices would be useful in the future proficiency tests.

This report focuses on the information received in the questionnaires and analysed the most relevant data about the participating organisations and technical aspects of the applied methods. The collected large amount of data gives possibilities of identifying best practices as well as pitfalls or erroneous practices that eventually can help laboratories to improve their methods and harmonise procedures.

Due to the vast amount of data in the REM 2020 PT, reporting is divided in three parts. The first part (Hult et al., 2024a) describes the PT and the results as stated above. A second report (Hult and Paepen, 2024b) describes outcome from the follow-up workshop and the third part (this report) describes answers from the survey.

2 Project management

2.1 REM2020 PT project coordination

The REM 2020 PT project was organised and coordinated by the Radionuclide Metrology team at the European Commission's Joint Research Centre (JRC-Geel, Belgium). The responsibilities amongst the involved staff of the organiser were defined at the project meetings. The JRC.G.II.6 unit of JRC-Geel is accredited for organisation of proficiency tests according to ISO/IEC 17043:2010.

2.2 Subcontractor

Some of the tasks (e.g. analytical work, production of PT test items/reference materials, PT data analysis) were subcontracted to other JRC-units and external expert institutes in the field already as part of the PT project. To process and analyse the large amount of additional information received from the on-line questionnaires, JRC-Geel subcontracted the Hungarian company "Social Organization for Radioecological Cleanliness", SORC (*Radioökológiai Tisztaságért Társadalmi Szervezet in Hungarian*). This organisation, founded in 1993, is an expert organisation in the NORM and environmental radioactivity measurement field.

2.3 Confidentiality and data protection

All data and results linked to the REM2020 PT were treated confidentially by the subcontractor, who only received anonymised results. Data treatment and privacy policy during the registration were compliant with the European General Data Protection Regulation (GDPR). For analysing the on-line survey affiliations or related personal information was not disclosed or shared by JRC with the subcontractor. The link between the participating organisations identities, on-line questionnaires and the submitted results cannot be identified by the subcontractor. However, it can be traced back by JRC-Geel staff only.

2.4 Data analysis and editing

The analysis of data in the questionnaires were done solely with Microsoft Office based software (MS Word and Excel). No Artificial Intelligence (AI) or AI based tools were used for data analysis and drafting text.

3 Analysis of the survey: information on the participating laboratories

The information in this chapter was provided in the questionnaires by the participants. In both questionnaires, on top of the contact data, participants had to fill in 50 fields by either answering multiple-choice or single choice questions or providing information in free-text. The free-text replies are all quoted exactly as given by the participants (e.g. including grammar errors). Some spelling errors might have been auto corrected by MS-Word and any information might have revealed the identities of the participants were deleted. For some questions (like what type of samples a lab measures) the authors have merged answers from various labs into lists under certain headings.

For some questions, selection of more than one answer was possible, which resulted in the sum of answers being higher than the total number of participants. The participants were also given the opportunity to give feedback and comments on the organisation of the PT. All relevant feedback will be considered in the future PTs. The PDF version of the blank registration- and reporting surveys span eight and ten A4 pages respectively (see Annex 1-2).

The replies were collated in spreadsheets where the registration- and reporting surveys included 6341 and 12204 data cells, respectively. These replies to questionnaires contain valuable, very detailed information about the laboratories and their methods.

3.1 Registration-survey

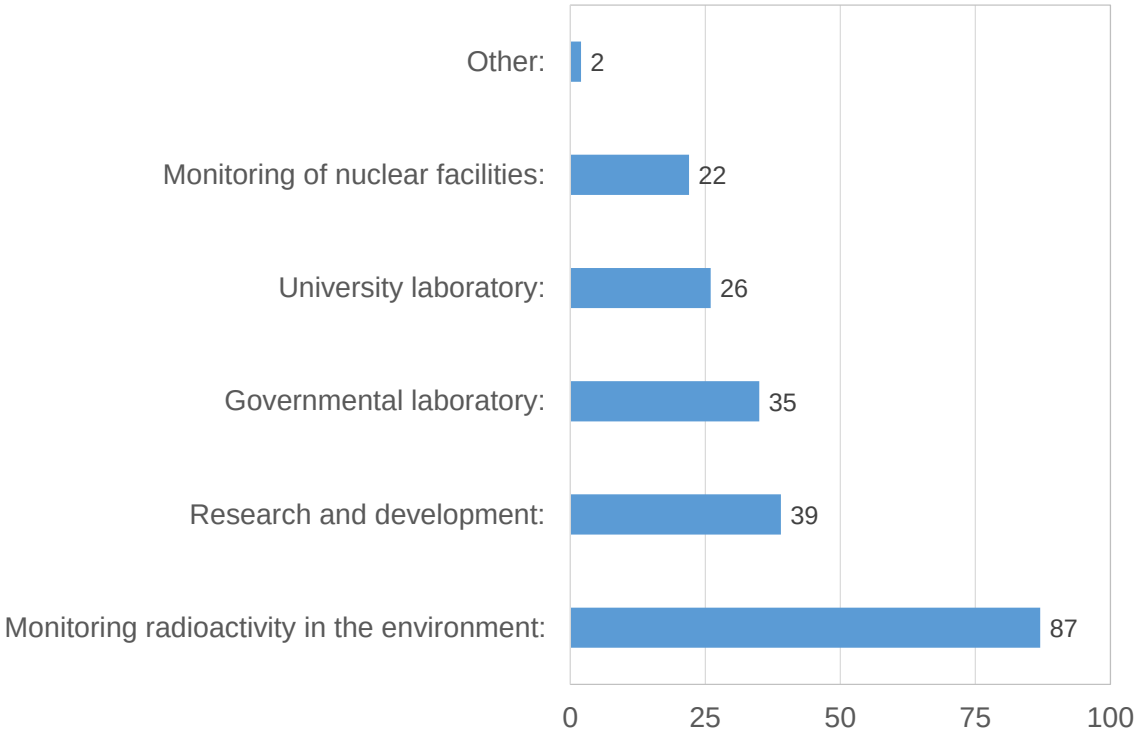
In total 108 organisations submitted the registration survey. Thus, JRC received 117 questionnaire files from them. As some laboratories already at the time of registration planned to apply multiple measurement methods, they submitted separate questionnaire files for each method.

3.1.1 Organisational details

Type of laboratory (Figure 1):

There are several different types of laboratories. Some are small and dedicated to one or two tasks. Others are big and in charge of monitoring, more or less, everything in a region. There are also laboratories that do occasional monitoring tasks or addressed for special studies. These facts need to be kept in mind this section. One reason for this is that there are so many different materials (matrices) to be monitored and so many different radionuclides. One laboratory cannot cover all possible combination of matrix/radionuclides.

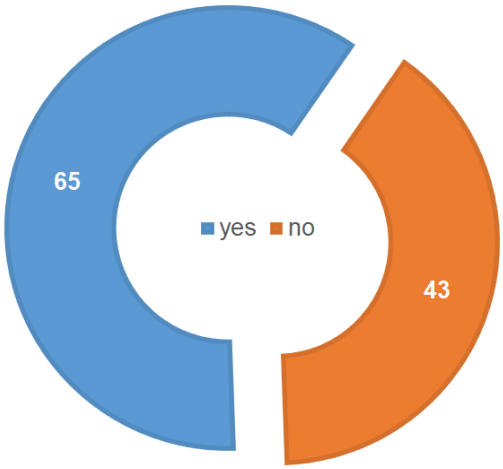
Figure 1. Number of different types of participating laboratories. (Multiple answers possible)



Source: JRC

Naturally occurring radionuclides in building materials are measured in the laboratory (Figure 2):

Figure 2. Naturally occurring radionuclides in building materials are measured in the laboratory.



Source: JRC

What kind of materials are usually measured in the laboratory?

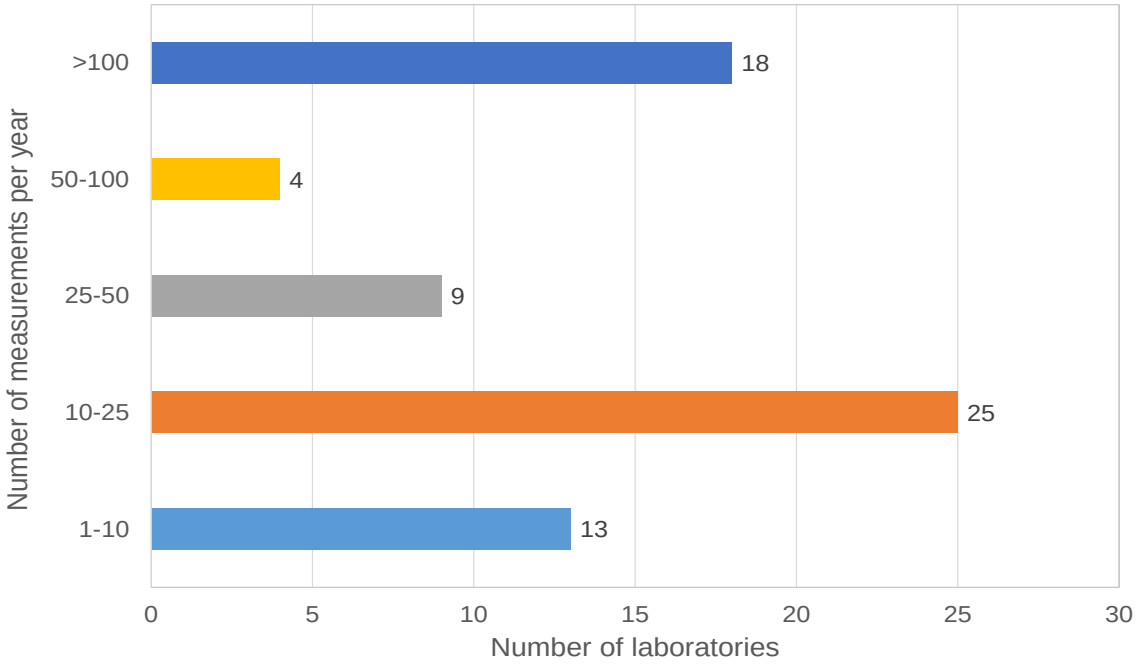
Various kinds of materials were mentioned by the participants, the majority is commonly available but there were some unusual examples as presented below (replies were copy pasted, just some minor typos were corrected). Answers were sorted into the five types below by authors at the time of writing this report.

- Natural building materials: geological materials, stone-based materials, sand, aggregates, gravel, stone, granites, limestone, tuff, marble
- Industrial building materials: concrete, cement, ceramics, glazes, pigments, bauxite, fluorspar, tiles, raw materials for cement- clinker
- (TE-)NORM[3] materials: raw materials, fly-ash, red mud, slag products, sludge, and phosphogypsum
- Environmental samples: wood, soil, sediments, food, seawater, saline solution
- Recycled materials: ballast, grout, plaster board, dust, loam, firebricks, dirt, alum shale, various debris, "all kinds"

Approximately, how many building material samples measured per year?

The number of building material measurements varies between 1 and 400 samples per year, with a median of 20 samples. Distribution of number of measurements and number of laboratories are plotted in **Figure 3**.

Figure 3. Approximate number of building materials measured annually.



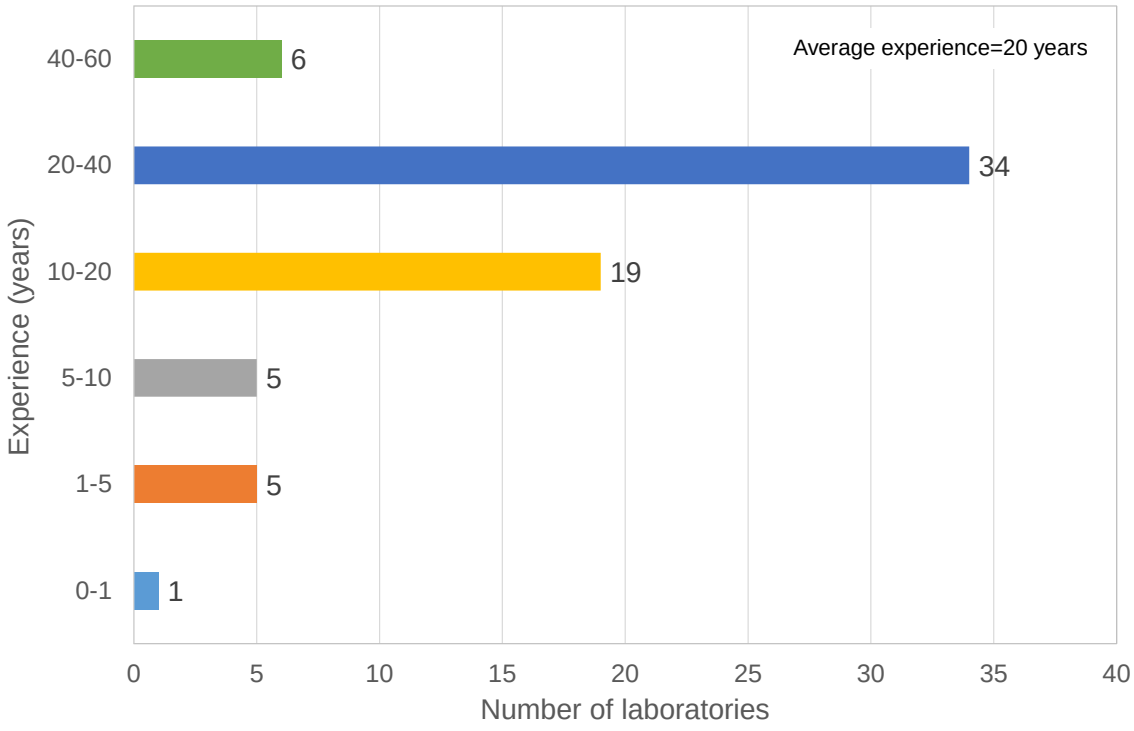
[3] TE stands for Technologically Enhanced. The term TENORM is less used today in favour of using the term NORM for all materials with natural radioactivity, no matter whether the activities are technologically or naturally enhanced

Source: JRC

How many years of experience do the laboratories have in performing this kind of measurements?

Years of experience varied between 0-60 years with an average and median of 20 years as seen in **Figure 4**. The most experienced laboratory performs this kind of measurements already for 60 years. On the other hand, there are laboratories that just started measuring construction/NORM materials in their laboratory at the time of the REM2020 proficiency test.

Figure 4. Years of experience in performing this kind of measurements.

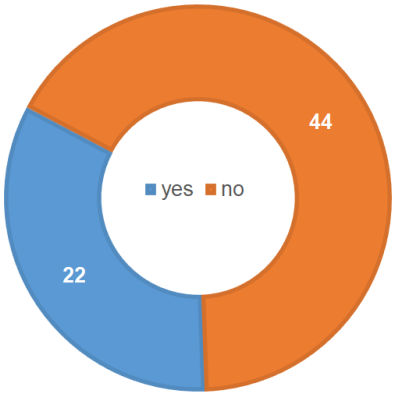


Source: JRC

Legally obliged to perform these measurements?

Figure 5 presents the number of laboratories that are legally obliged to perform measurements of building materials.

Figure 5. If the laboratory legally obliged to perform measurements of building materials.



Source: JRC

Only one-third of the participants that replied are legally obliged to perform these types of measurements either by regulatory bodies or ministries. Only few laboratories do it because of accreditation.

By whom?

— Regulatory bodies: 9 participants

— Ministries: 8 participants

— Accreditation bodies: 3 participants

— Other: 1 participant

The legal basis for this obligation

— National regulations, obligations: 16 participants

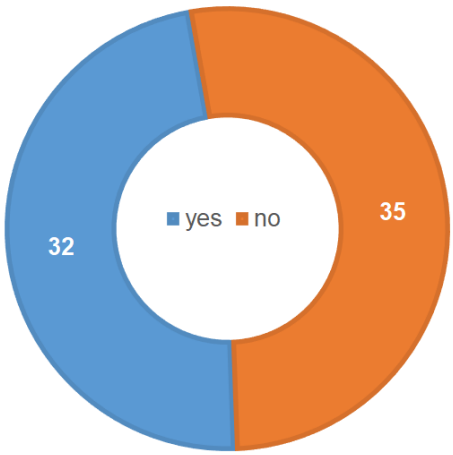
— Accreditation, ISO/IEC 17025: 3 participants

— Contracts: 2 participants

3.1.2 Samples and sample preparation

Do the laboratories perform moisture content determination in these samples (Figure 6)?

Figure 6. If the laboratories perform moisture determination and number of corresponding laboratories.



Source: JRC

What method do the laboratories use?

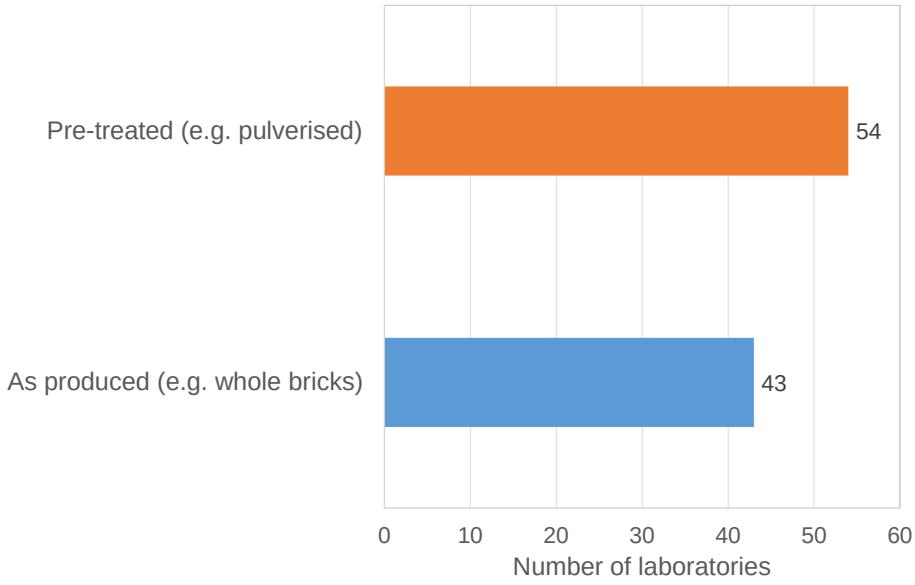
- In-house developed method: 21 participants
- Standard method (i.e. ISO, CEN or national standard): 9 participants
- Other: 5 participants

Reference to the standard used

- EN ISO 18589-3: 3 participants
- PN-EN 1097-5:2008 and PN-B-04481:1988
- IEC 61452:1995 Nuclear instrumentation - Measurement of gamma-ray emission rates of radionuclides - Calibration and use of germanium spectrometers
- ISO 20042:2019
- KRISS standard procedures
- LVS - 257:2000
- STN P CEN/TS 17216
- UNI 11665:2017
- IAEA Soil-327, Soil-6

In which form are the building materials received (Figure 7)?

Figure 7. Form of building material samples laboratories receive them and number of laboratories.

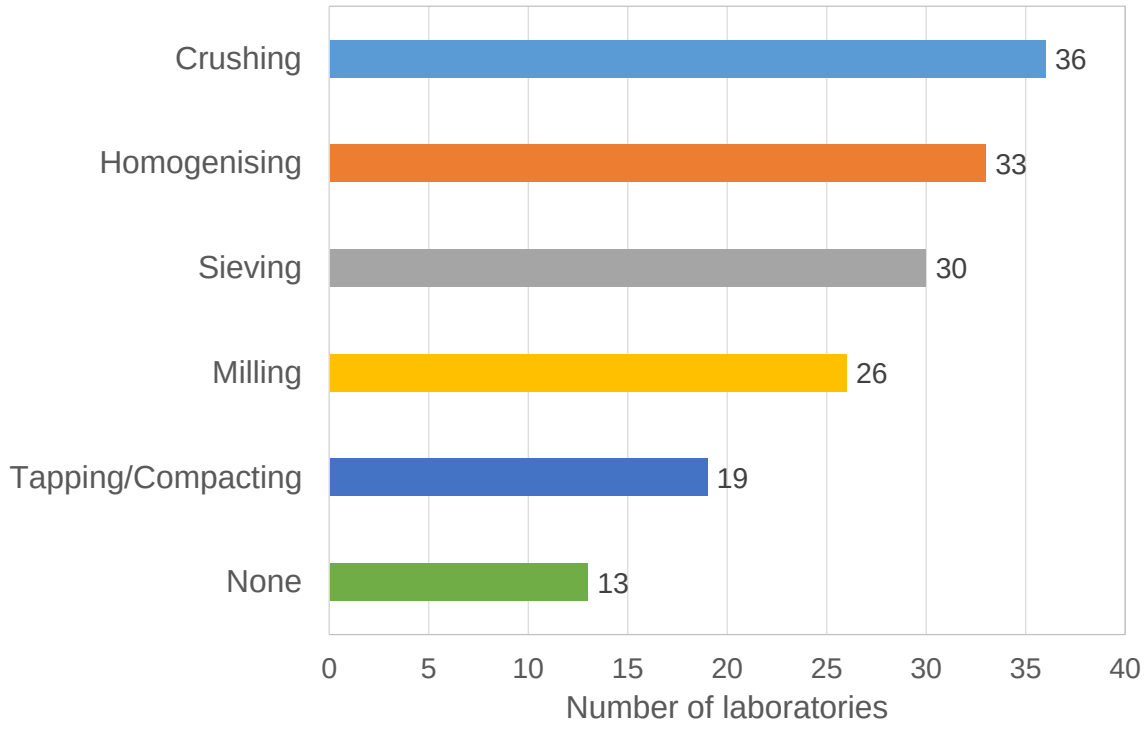


Source: JRC

3.1.2.1 What kind of processing is applied during sample preparation?

The frequency of the different processing approaches for sample preparation are presented in **Figure 8**.

Figure 8. Sample processing approaches and frequency of their use.



Source: JRC

Tools/apparatus used for crushing

- a jaw crusher, hammer, a commercial crushing machine, automatic crushing device, sledge-hammer, crusher/pestle, mill, manual crushing, presses and mortars, mallet, common household tools, manual crushing using a drilling machine, diamond saw,
- subcontracted if necessary to a specialized laboratory --> too high hardness.

Tools/apparatus used for milling

- ball mill, milling devices, Agate mortar, cutting mill, centrifugal mill, cross-hammer mill, vibratory disc mill (pulveriser), mallet, hoop mill, porcelain mortar, blender, laboratory blade mixer,
- subcontracted if necessary to a specialized laboratory --> too high hardness.

Tools/apparatus used for sieving

- a set of sieves, a commercial sieving machine, manual or automatic sieves, wet sieving, attested sieve, vibratory sieve shaker, CISA calibrated sieve.
- Manual sieving at 2 mm or 0.2 mm mesh depending of the analysis to performed

The mesh sizes of the sieves used

- 2 mm , 800 µm, 400 µm, from 0.02 mm to 100 mm, 4 mm; 2 mm; 250 µm;
- standard sizes of 63, 100, 150, 200, 250, 500 µm, 2 mm, 4 mm
- 2 mm (gamma spectrometry) or 0.2 mm (alpha spectrometry)

The typical most representative fraction, average and end-point used for sieving

- < 400 µm; 800 - 1200 µm
- 0-63 mm; 90 mm
- 2 mm; 4 mm
- <2 mm
- 63-200 µm, 4 mm for NORM
- <1 mm
- 200 µm
- 0/4 mm
- Gamma: 2-0.2 mm; Alpha: 0.2-0 mm
- “Different”/“Does not apply”/“I don't understand”
- “Typical faction of samples is hard to define. The methodology requires checking the condition of the maximum particle size of the sample. Sample should be crushing and milling until achieved maximum particle size < 2 mm.”

Tools/apparatus used for homogenising

- manual stirring, a commercial homogenizing machine, mixing with a spoon during several minutes, laboratory cuvette, laboratory mixer, drum hoop mixer, V-mixer, paddle, shredder, mixer, rotating blade homogenizer, Hammer- ball mill, tumbler, stir in a container
- Laboratory spatulas, spoons, spades, shovels

Tools/apparatus used for tapping/compacting

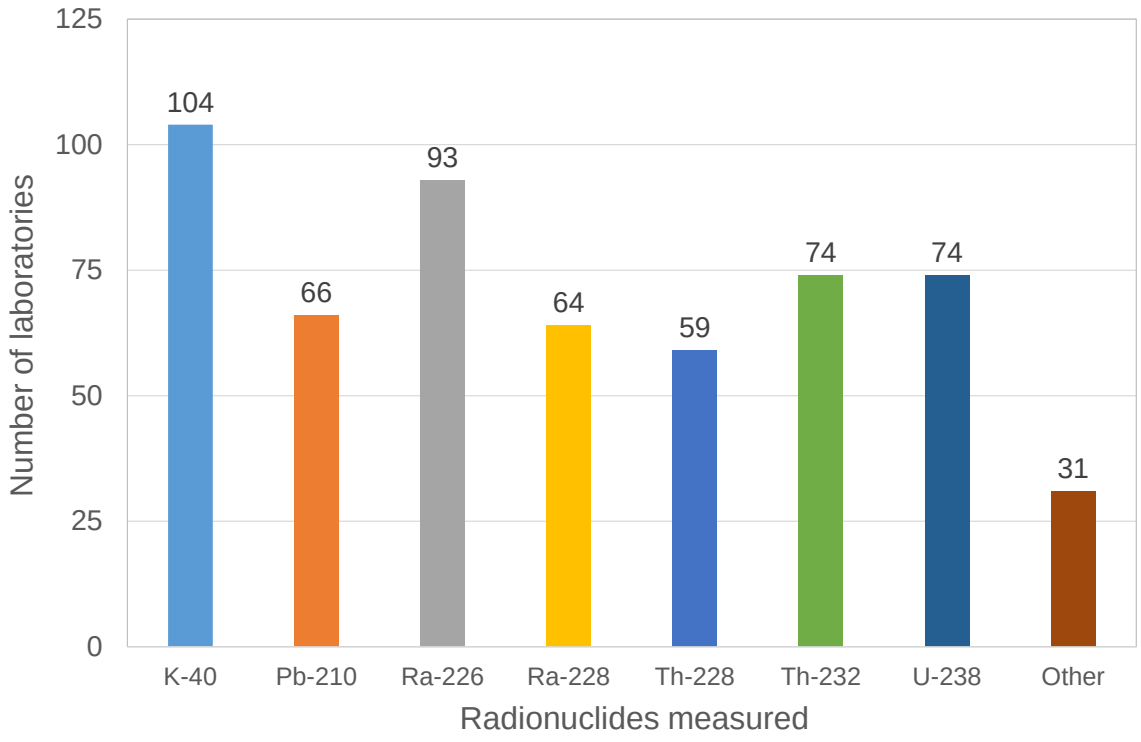
- manual stirring, silicone glow and plastic tape, compacting press, manually, proctor rammer/other rammer, laboratory spatulas, spoons, spades, shovels, manually with piston, home-made tools
- Compacting samples in measuring vessel by gentle tapping without using tools,
- If the material has very low density by hydraulic press,
- In-house manual cylindrical compaction tool adapted to the geometry of the urine bottle used as a sample container.

3.1.3 Measurements methods

3.1.3.1 Which naturally occurring radionuclides are routinely measured?

Apart from the seven requested radionuclides, participants routinely measure approximately 20 other radionuclides plus plutonium and americium isotopes as seen in **Figure 9** and the list under.

Figure 9. The routinely measured naturally occurring radionuclides and number of laboratories.



Source: JRC

Which other radionuclides…

— Be-7, Na-22, Co-60, I-131, Cs-134, Cs-137, Tl-208, Po-210, Pb-212, Bi-212, Bi-214, Pb-214, Rn-222, Ra-224, Ac-228, Th-230, U-234, U-235, Pa-234, Pa-234m, Pu, Am

3.1.3.2 Which method is used for the measurement of K-40?

— Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 102 participants

— Not applicable: 3 participants

— Other: 2 participants (ICP-MS)

3.1.3.3 Which method is used for the measurement of Pb-210?

— Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 62 participants

— Gamma-ray spectrometry with low resolution detector (e.g. NaI): 1 participant

— LSC: 1 participant

— Other: 2 participants (Radiochemical separation and measurement in a proportional counter; Bi-210)

3.1.3.4 Which method is used for the measurement of Ra-226?

— Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 88 participants

— Gamma-ray spectrometry with low resolution detector (e.g. NaI): 1 participant

— LSC: 1 participant

— Other: 3 participants; (1. proportional counter, 2. Gamma-ray spectrometry BGO crystal, 3. emanometry + solid scintillation)

3.1.3.5 Which method is used for the measurement of Ra-228?

— Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 63 participants

— Other: 1 participant; proportional counter

3.1.3.6 Which method is used for the measurement of Th-228?

— Alpha-particle spectrometry: 7 participants

— Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 51 participants

— ICP-MS: 1 participant

3.1.3.7 Which method is used for the measurement of Th-232?

— Alpha-particle spectrometry: 11 participants

— Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 54 participants

— Gamma-ray spectrometry with low resolution detector (e.g. NaI): 1 participant

— ICP-MS: 4 participants

— Other: 3 participants;
(1. Estimation from daughters, 2. Gamma-ray spectrometry BGO crystal, 3. MC-ICP-MS)

3.1.3.8 Which method is used for the measurement of U-238?

— Alpha-particle spectrometry: 12 participants

— Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 55 participants

— ICP-MS: 5 participants

— Other: 2 participants;
(1. alpha spec or gamma spec or LSC-depending on sample and purpose, 2. MC-ICP-MS)

Which method is used for the measurement of additional radionuclide(s), not listed in the first question?

— Alpha-particle spectrometry: 4 participants

— Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 26 participants

— ICP-MS: 1 participant

3.1.3.9 Brief description of the sample preparation, if any (gamma-ray spectrometry)

Some typical examples of sample preparation steps are listed below. The more detailed information from the participants is presented in Annex 1.

— as it is (no preparation)

— depends on the sample type

— 1/ crushed and milled if necessary, 2/ put in a container, 3/ gas tightness bag, 4/ +/-30 days waiting time, 5/ measurement.

— drying to constant mass (some labs specified the drying temperature, 105 °C)

— weighing, drying

— weighing into a Marinelli container (0.5-1L) and waiting for radium-radon equilibrium (14-20-30 days) then measured

— shaking and homogenization

— Crushing, homogenisation, weighing

— Drying, sieving, crushing, milling, homogenising and reducing, packing in a measuring vessel.

— addition of charcoal powder to trap Rn in the sample volume.

— Using sealed containers,

— an aliquot of the sample is placed in a measuring geometry - the sample container is filled up to the top and then sealed inside an aluminium foil

— After sieving to 2mm. sample packed in a plastic bottle of standard geometry (100ml, 250ml or 500ml). Addition of 10% by volume of activated carbon during the preparation. The measurement of Ra-226 carried out after 30 days of equilibration.

— Samples are oven dried, sieved through a 2mm sieve and homogenised. An aliquot is placed in a well-defined counting geometry and then measured on a high-resolution gamma spectrometer. Appropriate corrections (TCS and self-attenuation) are applied.

— Sample drying at 40°C, sieving through 2mm and drying at 110°C. Another step: sieving through 0.5mm and then measuring by gamma-spectrometry.

— According to - STN EN ISO 18589-3 standard

3.1.3.10 Brief description of the sample preparation, if any (destructive methods)

— Spiking tracers, dissolving in a mixture of acids, chromatographic separation

— ashing at 600 °C and acid digestion

— alpha source preparation: electrodeposition or micro coprecipitation

— liquid scintillation counting: liquid/liquid extraction by selective cocktail

As observed from the survey, there were several different sample preparation approaches for the gamma-ray spectrometry. On the contrary, the basic sample preparation steps for the destructive analytical methods (ICP-MS, alpha-spectrometry) were more harmonised among the laboratories.

3.1.3.11 Which type of efficiency calibration do you use for gamma-ray spectrometry?

— Reference material or source: 63 participants

— Monte Carlo calculations only: 7 participants

— Combination of Monte Carlo calculations and reference material or source measurements: 28 participants

— Other: 5 participants (1- multi-gamma source, 2- LABSOC programme from Mirion technology, 3- ISOCS, 4- in-house developed, 5- mathematical calculation)

Is the measurement container for gamma-ray spectrometry completely filled with the sample?

— Yes: 68 participants

— No: 35 participants

Are there any fillers applied to fill the void?

— Yes: 2 participants

— No: 33 participants

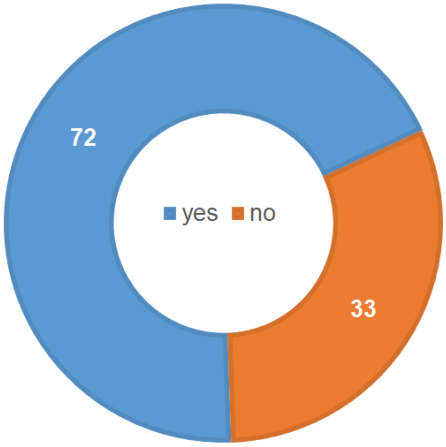
Please provide additional information on the type of fillers used

— Marinelli one litre and one-half litre,

— The void is filled with paraffin and when the paraffin is still warm the jar is closed airtight.

Do you apply any sealing material to close the measurement container for gamma-ray spectrometry (Figure 10)?

Figure 10. Number of laboratories apply or not any sealing material to close the measurement container.



Source: JRC

What kind of sealing it do you apply to close the measurement container?

— Bag

— silicone glue and plastic tape

— Lid and plastic bag

— PVC tape

— wrap with Teflon tape

— vacuum packing

— Lukopren T1990

— One-component silicone permanent plastic sealant.

— Dispense Parafilm

— plastic insulator

— Scotch tape (seal)

— Samples are usually sealed with epoxy glue,

— Tape around the lid

— Marinelli geometries with double lid and Parafilm

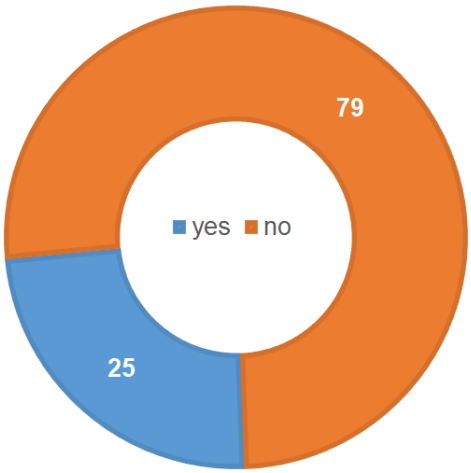
- adhesive tape
- sealing with rubber tape
- Paraffine film
- Sealed plastic bags not permeable to radon
- Special lab tapes with wax
- Container is sealed in a radon-proof plastic bag.
- wrapping in a soldering material
- insulating elastic duct tape
- Tape wrapped around
- lift tap sealed with a paraffin film to avoid radon leakage
- An external can
- Thermic glue
- insulating tape
- To seal the measurement container we use the scotch tape and a plastic bag
- silicone
- Parafilm
- The sides of the lid of the metal container are covered with a layer of plasticine (modelling clay) which is covered with a layer of isolation tape.
- The containers are sealed with tape.
- Duct tape
- in-house developed method
- a vacuum machine to seal the samples inside an aluminium foil (PET/aluminium layer)
- Activated carbon for trapping radon and daughter products
- for Ra-226 measurements, completely filled steel containers with gas proof lid are employed
- "If applicable (depending on sample material - no powder), completely filled sample containers (material covered with a plastic disk) are vacuum packed into layered PA / PE bags;
- In case of a powder-like material, aluminium bottles with PE cap and PTFE gasket are used."
- Sanitary silicone
- aluminum tape
- wrap the container with Teflon or masking tape
- Petri disks are sealed with silicone gel around the borders. After 24h drying, Petri boxes are packaged in vacuum plastic bags.

- paraffin
- A sticky tape.
- Resin
- O-ring
- adhesive tape
- Vacuum packaging of the sample container in a sealed aluminium lined bag.
- Radon tight plastic and vacuum packer.
- silicon tape

Do you test the radon tightness of the measurement containers used for gamma-ray spectrometry?

Replies on whether laboratories test or not for radon tightness of the measurement container are presented in **Figure 11**.

Figure 11. Number of laboratories test or not for radon tightness of the measurement container.



Source: JRC

Please provide a brief description of this method

- if needed we put on the top plastic cower and clime with glue
- Enclose the container of the sample in a 100 L metal tight vessel and Rn leakage from the sample in the vessel air measured using a Lucas cell counting device.
- Observation of the decay curve of radon in a high radon activity water sample. The deviation of the experimental value of the decay constant allows the quantification of the leakage
- Internal method
- Reference sample in the same container is measured in a airtight container.
- All the radiometric measurements were performed by low-level high-resolution gamma spectrometry with coaxial-type germanium detectors (Canberra Industries Inc., USA). These detectors are enclosed in 15 cm old iron shields inside a clean laboratory. The laboratory is kept under positive pressure to minimize the concentration of radon in the indoor air. These detectors were calibrated using certified reference gamma-ray cocktails purchased from the National Physical Laboratory (United Kingdom). The laboratory measurements are subjected to a regular quality control program including periodical monitoring of the backgrounds, calibrations, and the detection system performances
- A sealed container is used to control the radon tightness of the beaker. The radon concentration in the sealed container, measured with an Alphaguard device, is an indicator of the tightness of the beaker.
- in-house developed method

- So far we have only tested the radon-tightness using a reference material of our national metrology institute. We currently have a project underway that will test the radon-tightness using radon-rich water (500Bq/l).
- Validated during the method validation (ISO 17025 accredited for Ra-226 analysis)
- attainment of secular equilibrium with Ra-226 sources, measurement of certified reference materials (Ra-226 measured by Rn-222 progenies), intercomparisons (Ra-226 measured by Rn-222 progenies)"
- Aluminium bottles are filled with water from a natural spring (containing about 500 Bq/L Rn-222), measurements on same HPGe detector are repeated for several days to check whether the decay-corrected Rn-222 concentration (determined via Pb-214) remains constant. For the method using vacuum packing we have an ongoing project to test radon-tightness using water from the natural spring described above.
- We use a RAD7 radon monitor in sniff mode around the sealed containers.
- By the method of accumulation in a closed chamber
- measurement by gamma-ray spectrometry at different equilibrium times
- Measurement of 186 keV peak (direct Ra-226 after deconvolution with U-235 peak) and afterwards, comparison with Pb-214-Bi-214 activity concentration. U-235 is measured by either gamma or alpha spectrometry to cross-check the right U-235 subtraction.
- with reference materials and spiked water
- We have studied the radon tightness of the Marinelli containers with water samples with different sizes and materials. We have measured these samples during several days and we have computed the radon diffusion per day.
- use of standard containing Ra-226
- Intercomparison measurements of radon in water by gamma spectrometry and emanometry measurements (ISO 13164-3)
- https://doi.org/10.1016/j.jenvrad.2019.05.007
- Vacuum packaging of the sample container in a sealed aluminium lined bag.
- We make sure that the daughters of Ra-226 are in equilibrium before we report Ra-226
- after the container is closed, the air is removed from the container by hand pushing

Please describe the calibration procedure(s) used for your measurement method(s) (other than gamma-ray spectrometry)

- isotope dilution analysis
- External calibration using reference solutions and/or use the spike as a reference and run samples in standard bracketing
- For the proportional counter measurement, a verification is made in efficiency and spillover with standard tables of Am-241 and 90Sr/90Y.

- For alpha spectrometry, the equipment is calibrated with a triple alpha steel plate of U- 233,Pu- 239+240 and Am-241 of certified activity
- Calibration is make using reference standards of U and Th. We also use Iridium like Internal Standard
- Isotopic dilution method
- NORM IAEA-148 with direct traceability - and density
- Alpha-particle spectrometry: Determination of counting efficiencies at different sample-detector distances, using certified radioactive sources.
- ICP-MS: Pre-adjustment of measurement parameters using uranium standards, use of certified material in each series of measurements and post-measurement corrections.
- CRM and specific source (peak to peak) applied for calibration (Energy and Efficiency).
- MCNP applied for self-attenuation correction and coincidence summing effect
- preparation of sources in different geometry (similar to samples) with known activity
- our extraction method for alpha emitters uses a tracer standard to determine the chemical yield
- For alpha spectrometry we used tracers of known activity
- gamma-ray spectrometry
- U-238 analysis (ICP-MS); The determination of metals by inductively coupled plasma mass spectrometry (ICP-MS) is performed by quantitative determination after calibration with suitable calibration solutions. The relationship between signal intensity and mass concentration is usually a linear one over at least five orders of magnitude. The ICP-MS is tuned daily by running the daily performance check solution and the “Smart Tune Daily” wizard. The QID STD/DRC is run followed by the Daily Performance Check and if that fails, a full optimisation is performed using the Smart Tune Daily wizard. On completion, verify that the calibration curve is acceptable for all reported elements – correlation coefficient should be > 0.995 and that blank results are < LOQ in each case. For metals whose calibration extend above 2 million cps, the linearity between the pulse and analogue measurements should be verified visually i.e. data from the highest pulse measurement should lie on the same calibration line as the point from the highest analogue measurement – if they do not, this may suggest that a Dual Detector Calibration may need to be performed.
- Use of standard Materials of reference
- alpha spec: radiochemical procedures: radioactive tracers are used
- LSC: validation tests with traced materials (U-236)
- Pb210 by proportional counting: stable carriers are used"
- Alpha spectrometry; Reference material.
- If required, self-absorption corrections are applied.
- TCS is also applied if required although natural radionuclides are included in the calibration source.

— Isotope-dilution with Po-209

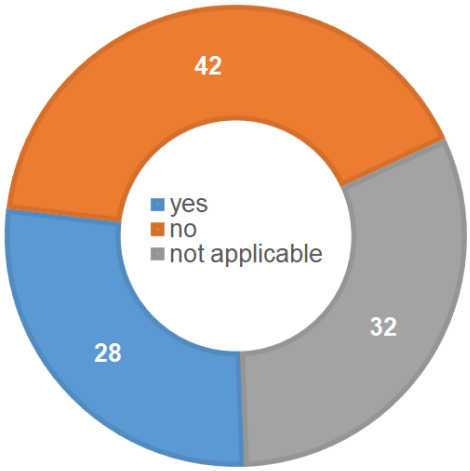
— Use of suitable tracers for alpha spectrometry.

— Use of Pb-210 ad-hoc calibration source for proportional counter.

3.1.3.12 Any difficulties while measuring the naturally occurring radionuclides in building materials?

The number of laboratories experiencing difficulties in measuring building materials are presented in **Figure 12**.

Figure 12. Experiencing any difficulties while measuring building materials.



Source: JRC

Description of difficulties

Text was copy-pasted from the questionnaire, as introduced by the participants (including typos and grammar errors).

— Availability of CRMs

— Standard used in not in the matrix

— For some radionuclides (Ra-226) problem of the deconvolution of the peaks; sensitivity problem for Pb-210 (self-absorption); disequilibrium between radionuclides belonging to the natural chains

— Sometimes we had problems with an apparent disequilibrium of the natural radioactive chains. Low gamma energy radionuclides such as Pb-210 are also challenging issue

— The samples need to be pre-treated, grounded

— Preparation of the samples

— Absorption coefficients at low gamma energies retrieval, use or determination

— measurement of Ra226 due to escaping of radon

- measuring Ra-226 using Rn-222 daughters without checking leakage tests has important uncertainties.
- The most critical issue is related to auto-absorption corrections for materials characterised by heavy metals; efficiency calibration for these materials can be strongly affected and the reliability below 100 keV is critical.
- the method in alpha spectrometry with the use of the tracer does not determine the real chemical yield, but appropriate correction coefficients must be applied
- Material density and composition, equilibrium in the natural radioactive chains
- If there is not enough material to fill the beaker up to the top we search the way to fill the container to prevent Rn escape and efficiency change
- Calculate activities with disequilibrium in the natural chains
- we are planning to purchase an vacuum closing apparátus to achieve the equilibrium between daughter elements.
- Correction must be applied for density and self-absorption.
- Variability while measuring background levels.
- radon tightness.
- determination of chemical composition of sample (for Monte Carlo modelling)"
- The time required as we leave prepared samples to stand for 30 days before analysis so the Pb-214 and Bi-214 reach equilibrium.
- Pb-210 can be difficult to accurately measure as it has a low energy line at 46Kev and will be affected by self-attenuation.
- For about 5% of samples the seal of the aluminium foil is faulty and needs to be re-applied (this is very visible as the foil expands when the vacuum is broken)
- High active samples to manipulate in a environmental laboratory.
- Toxicity of the industrial samples.
- Waste management after analysis
- Gamma spec: self-absorption management; subsamples extraction, representativity and homogeneity
- Radiochemical procedures: sample dissolution; subsamples extraction, representativity and homogeneity"
- Sometimes the elemental composition is not known (then it is difficult to determine the Pb-210 content precisely)
- Sometimes it is difficult to crush the material uniformly by hand (e.g. firebricks)
- Doubts about the correct closure of the container to avoid radon losses
- Determination of Ra-226 from 186 keV emission with interference corrections

— Availability of reference standard materials of high density for efficiency calibration"

— the samples are too active for preparation in a white zone, you never know in advance what's in the sample

— risk for contamination of material: mill, sieves, ...

— difficult to mill, we then do the material to the central technical services

— The density of the sample, the calibration source, the radon generation in the void, and so on. It is an interesting and actual issue. There are a lot of difficulties described in the biography.

— In case we receive the material not already prepared, as a bulk material.

— Autoassorbimento, affetto somma ed omogeneizzazione di particolari campioni. (translation by JRC/Authors: Self-absorption, summation and homogenization of particular samples.)

3.2 Reporting-survey

There were 85 laboratories that submitted 89 questionnaire files where some of the questions were repeated just like in the registration survey. Participants were asked whether they changed something in their standard procedure they introduced in the registration-survey. If so, then they were requested to elaborate those changes and give the main reasons for doing that. The willingness to provide information was in general high but for certain questions it was low, sometimes less than 50% of the participants provided information.

3.2.1 General information on the performed measurements

Did you follow your standard procedure during the measurement of the PT reference materials?

— Yes: 85 participants

— No: 0 participant

Did you determine the moisture content of the samples according to the provided procedure?

— Yes: 81 participants

— No: 4 participants

What type of changes did you make?

— The sample was dried in an oven at 105°C to constant weight as the activity concentration was requested in Bq/kg dry mass.

— The material was tested in its delivered form

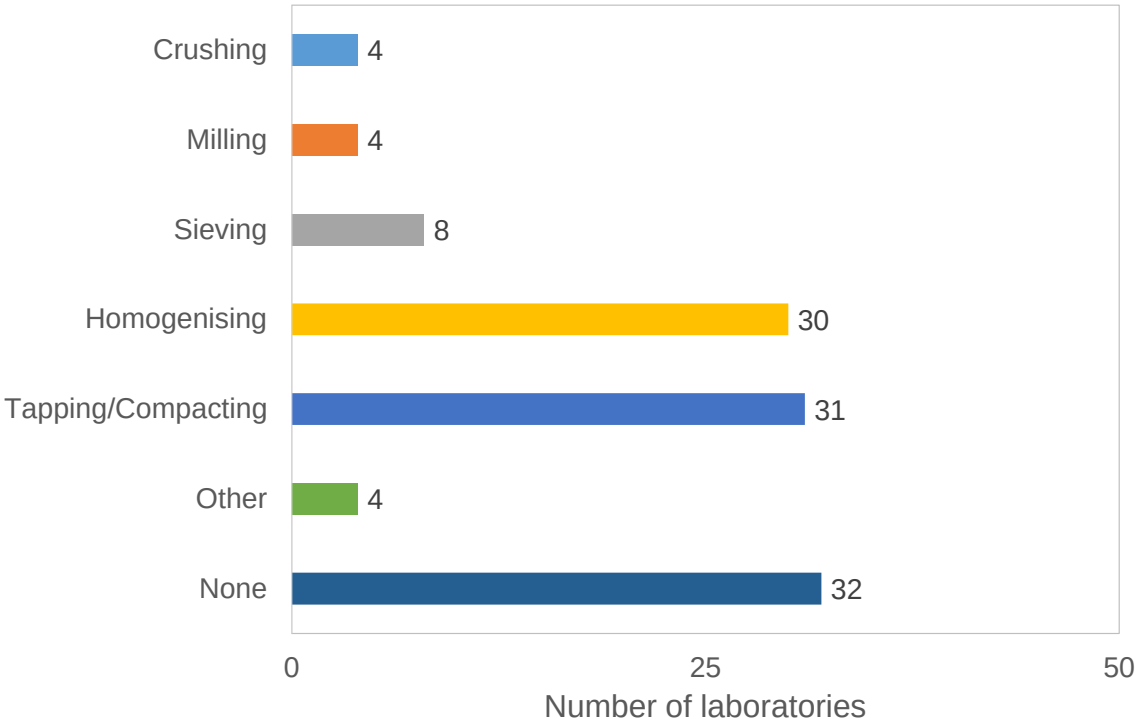
— One sample was measured

— Our standard procedure of drying the sample were followed, weighing before and after.

3.2.1.1 What kind of processing did you apply during sample preparation of NORM1?

The different processing procedures for NORM-01 sample preparation are presented in **Figure 13**.

Figure 13. Processing procedures for NORM-01 sample preparation.



Source: JRC

More information:

— An epoxy resin was used to shield the container for radon.

— the dried samples were measured as such

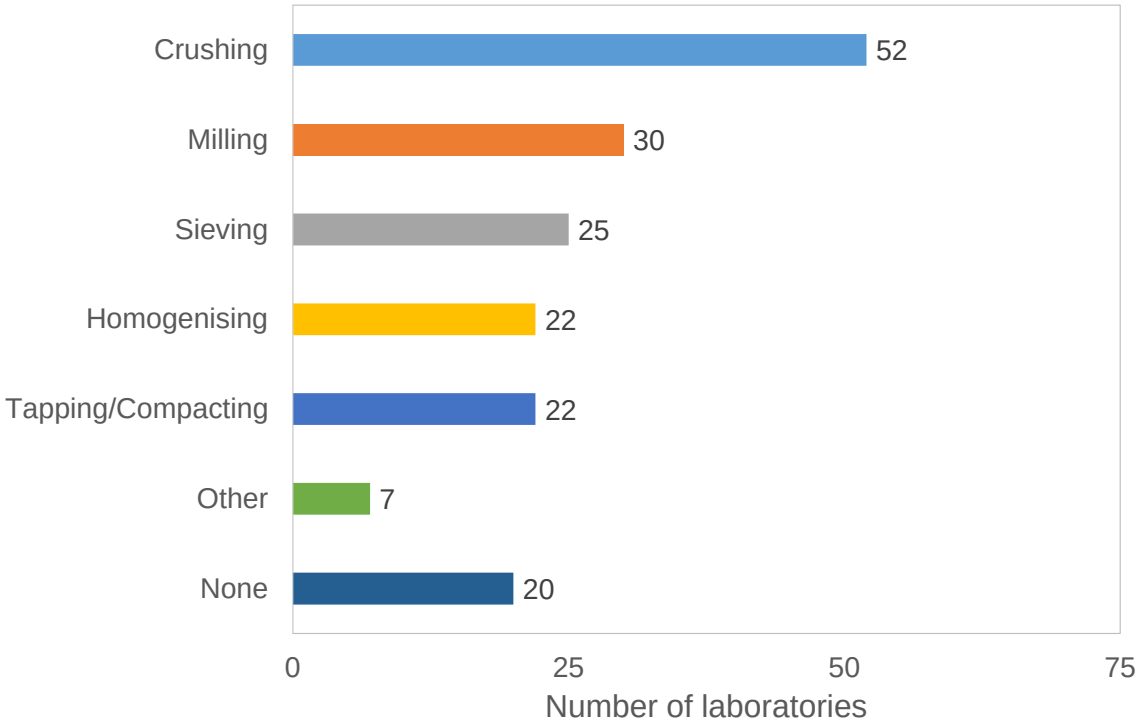
— hermetization

— drying

3.2.1.2 What kind of processing did you apply during sample preparation of NORM2?

The different processing procedures for NORM-02 sample preparation are presented in **Figure 14**.

Figure 14. Processing procedures for NORM-02 sample preparation.



Source: JRC

Tools/apparatus used for crushing

— hammer, ball mill, manual tools, milling machine, a jaw crusher, hammer and chisel, standard inox grinds, stone grinder, manual crushing using a drilling machine, pestle in ceramic bowl, jam tooth crusher, crushing machine, mortar; sample is cut and then Retsch crusher.

Tools/apparatus used for milling

— mortar, Retsch fine miller, jar mill machine, ceramic mill, mortar grinder, ball mill, vibratory disc mill (pulveriser), vibrating cup mill, Agate mortar, magnetic stirrer, hammer crusher, milling using a W-miller, porcelain mortar

Tools/apparatus used for sieving

— a set of sieves, certified sieve, vibrating sieve, square mesh sieves

The mesh sizes of the sieves used

— 75-125-200-250-400-500 µm

— 0.5-1-2-3-4-5-6 mm

— <2 mm

— first step 5 mm, second step 2 mm, third step 1 mm

Tools/apparatus used for homogenising

— manual mixing in the container, press, with spatula, manual shaking, steel pallets and steel container, shovel, rotating drum, blender, electric homogenizer, stirrer, bucket, spoon

— shake in a large bowl, then divide in subgroups, and lastly regroup. Repeat this at least 3 times.

Tools/apparatus used for tapping/compacting

— manual, laboratory spoon, hand compacting, tapping the container on the bench/table, a press machine, home-made tool, dental vibrator, piston, hydraulic press with piston, centrifuge, vibrator table, spatula and pestle, spoon, silicon gel + Plastic tape

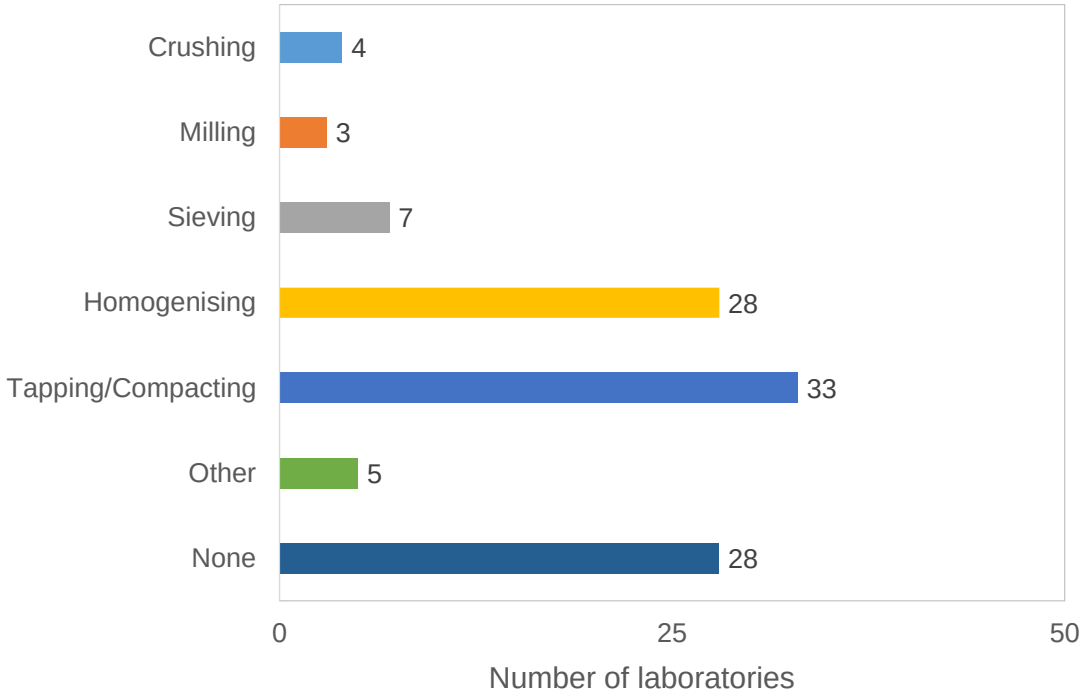
— other: drying, grinding, 1 piece was cut from the whole block and measured.

— Not able to apply the routine procedure

3.2.1.3 What kind of processing did you apply during sample preparation of NORM3?

The different processing procedures for NORM-03 sample preparation are presented in **Figure 15**.

Figure 15. Processing procedures for NORM-03 sample preparation.



Source: JRC

More information:

— An epoxy resin was used to shield the container for radon.

— A cylindrical sample was drilled out of the bulk material.

— the dried samples were measured as such

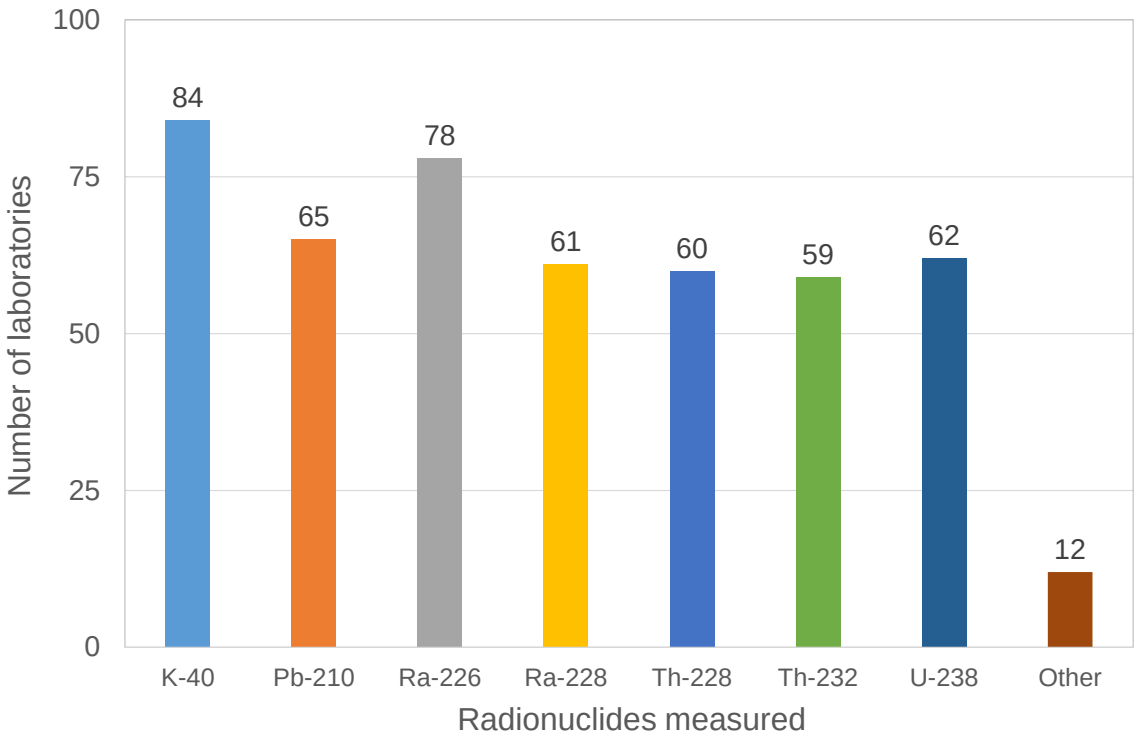
— hermitization

3.2.2 Measurements methods

Which naturally occurring radionuclides did you measure in the REM 2020 PT reference materials?

The different naturally occurring radionuclides measure in the REM 2020 PT are listed in **Figure 16**.

Figure 16. Naturally occurring radionuclides measured in the REM 2020 PT reference materials.



Source: JRC

Which other radionuclides...

- Tl-208, Po-210, Pb-210, Pb-212, Bi-212, Bi-214, Pb-214, Ac-228, Th-234, U-234, Pa-234m
- Th-230 (gamma sp., alpha sp.), U-234 (alpha sp.), U-235 (gamma sp., alpha sp.), Ac- 227 (gamma sp.)

Which method was used for the measurement of K-40?

- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 79 participants
- Gamma-ray spectrometry with low resolution detector (e.g. NaI): 3 participants
- By determination of total potassium: 1 participant
- Other: 1 participant (BGO detector)

Which method was used for the measurement of Pb-210?

- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 64 participants
- LSC: 1 participant

Which method was used for the measurement of Ra-226?

- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 73 participants
- Gamma-ray spectrometry with low resolution detector (e.g. NaI): 2 participants
- LSC: 1 participant
- Other: 1 participant (HPGe, equilibrium conditions, derived from Pb-214)

Which method is used for the measurement of Ra-228?

- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 60 participants
- Other: 1 participant; (HPGe, equilibrium conditions, derived from Pb-214)

Which method is used for the measurement of Th-228?

- Alpha-particle spectrometry: 5 participants
- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 54 participants
- Other: 1 participant (HPGe, equilibrium conditions, derived from Pb-212)

Which method is used for the measurement of Th-232?

- Alpha-particle spectrometry: 12 participants
- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 39 participants
- Gamma-ray spectrometry with low resolution detector (e.g. NaI): 3 participants
- ICP-MS: 1 participant
- Other: 4 participants;
  (1. HPGe detector, equilibrium conditions, derived from Ra-228, 2. estimated as ma Th- 228, Ra- 228, 3. ICP-OES, 4. HPGe detector, assumed that Th-232 decay products are in equilibrium)

Which method is used for the measurement of U-238?

- Alpha-particle spectrometry: 12 participants
- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 46 participants
- ICP-MS: 1 participant
- Other: 3 participants;
  (1. U-238 is assumed to be in equilibrium with the Pa-234m and Th-234, 2. ICP-OES, 3. deconvolution U-235/Ra-226 (Ra-226 derived from Pb-214); using factor 21.73 U-238/U-235)

Which method was used for the measurement of additional radionuclide(s), not listed in the first question?

- Alpha-particle spectrometry: 2 participants
- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe): 10 participants

3.2.2.1 Brief description of the sample preparation, if any (gamma-ray spectrometry)

Some typical examples of sample preparation steps are listed below. The more detailed information from the participants is presented in Annex 1.

— No sample preparation: 17 participants (NORM 1 and NORM 3)

— Moisture determination: 3 participants

— Homogenising only, before filling the test portion in the measuring geometry: 7 participants

— Drying and homogenising: 8 participants

— Processing (e.g. sieving, milling crushing, compacting): 32 participants

— Closing measurement geometry radon-tight: 21 participants

After sample preparation, samples were stored until equilibrium reached. For the storage time only limited number of participants gave concrete information (3-4 weeks: 5 participants; min. 14 days-30 days: 8 participants) but at the other section more extended feedback was received from the participants.

3.2.2.2 Brief description of the sample preparation, if any (alpha-particle spectrometry, ICP-MS, LSC)

— Alpha-spectrometry: total digestion of samples with acids followed by chemical separation.

- Ashing or microwave digestion using pure acids or acid mixtures (HF, HF+HNO_3 or HF-HNO_3-HCl-H_2O_2) or fusion (NaOH)
- U-232 and Th-229 yield tracers
- Pre-concentration of actinides: dissolution of dried residue, $Fe(OH)_3$ coprecipitation or liquid-liquid solvent extraction by using tributyl phosphate (TBP)
- Separation/purification on anionic exchange resin and extraction chromatographic resin
- Alpha source preparation by NdF_3 co-precipitation or electrodeposition

— ICP: the samples were digested with a mixture of 2.5 M HNO_3 and 0.5 ml H_2O_2 at 90 °C overnight in clean lab conditions.

— LSC: fusing of 1 g sample with NaOH. No more details were shared on the procedure, cocktails.

What was the volume of the measurement container for the gamma-ray spectrometric measurements? (in litres)

— 0.061-3.375 L

What was the mass of the sample used for the gamma-spectrometric measurements? (in grams)

— 0.1-3777 g (average 414 g)

What was the mass of the sample used for the measurements using alpha-particle spectrometry? (in grams)

— 0.1-10 g (average 2 g)

What was the mass of the sample used for the measurements using ICP-MS? (in grams)

— 5 g

What was the mass of the sample used for the LSC measurements? (in grams)

— 1 g

Which type of efficiency calibration did you use for gamma-ray spectrometry?

— Reference material or source: 65 participants

— Monte Carlo calculations only: 15 participants

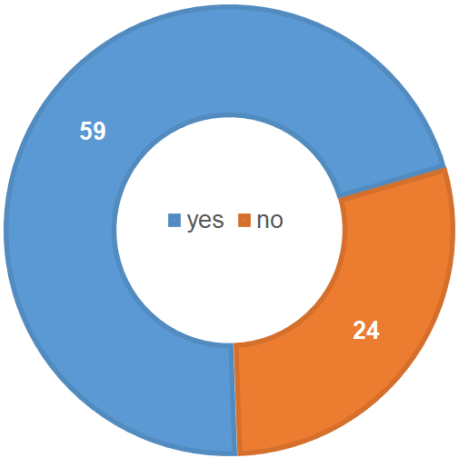
— Combination of Monte Carlo calculations and reference material or source measurements: 14 participants

— Other: 3 participants:
(1- IAEA-RL-148 reference material, 2- LABSOC programme, 3- Canberra ISOCS Programme)

Were the measurement containers for gamma-ray spectrometry completely filled with the sample?

Information if the measurement container was completely filled with the sample is in **Figure 17.**

Figure 17. Number of cases when measurement containers for gamma-ray spectrometry completely filled with the sample.



Source: JRC

Were there any fillers applied to fill the void?

— Yes: 3 participants

— No: 21 participants

Additional information on the type of fillers used

— Expanded polystyrene foam

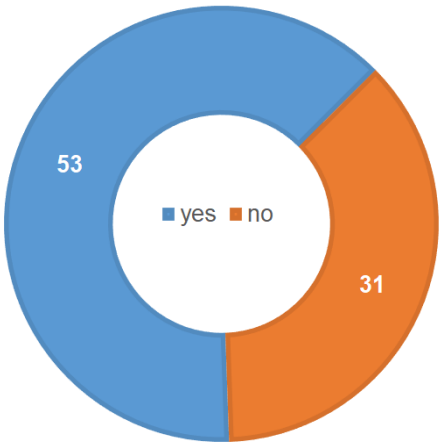
— sand with very low radioactivity

— paraffin was poured over the sample and then the vial was closed, and sealed with the paraffin

Did you apply any sealing material to close the measurement container for gamma-ray spectrometry?

Information on applying sealing materials to close the measurement container is in **Figure 18.**

Figure 18. Number of cases when sealing material was applied to close the measurement container for gamma-ray spectrometry.



Source: JRC

What kind of sealing did you apply to close the measurement container?

— Epoxy resin or glue

— silicone + tape

— Plasticine covered with isolation tape

— containers with auto-sealing cover

— Parafilm

— Vacuum sealing in aluminium box container

— Tapes: adhesive tape, duct tape, aluminum tape, insulating tape

— Sealed by parafilm and a radon-proof plastic bag

— Silicone and plastic bag using vacuum system

— Vacuum sealed bag

— wrapped by Al foil

— Baysilone paste

— canning

— Metal (Al) Marinelli beaker with “O”-ring

Description of the calibration procedure(s) used for your measurement method(s) (other than gamma-ray spectrometry).

— ICP-OES calibration lines prepared in 5 different concentration points by using the related standards.

— Alpha spectrometry: radiotracers (certified reference material) were used

— Use of mixed source with three alpha emitters (U-233, Pu-230+240 and Am-241)

— A soil was traced with different artificial radionuclides and Pb-210 and placed in a container for the gamma-ray spectrometric measurement.

— Efficiency calibration for PIPS detector in order to determine the chemical yield of the process. Energy calibration of PIPS detectors.

— By using reference materials, sources

— internal tracer method for U-238 and Th-232 alpha analyses, tracers by NIST, home diluted

— spiked samples of the same material were used in tandem with ordinary sample. Alpha spectrometry was performed with Th-229 and U-232 spikes

— ICP-MS: six-point linear calibration using standard solution

— Ver cubes of K, U and Th made of IAEA RL 148 reference material

3.2.2.3 Detection limits of the radionuclides measured.

The reported detection limits for radionuclides and materials are collected in **Table 1**. Analysing the reported detection limits, it was noted that some of the participants introduced a large range as detection limits which makes difficult to assign a detection limit value. In one case for all three PT materials the following detection limits were submitted: 15- 1000 Bq/kg and 50-5000 Bq/kg for Ra-226 and K-40 respectively. These data were not used for further analysis since it seems very unrealistic to have detection limits in the range of few hundreds of Bq/kg. Reporting such uncertainties is a clear indication of some issues, e.g. either the method is not under control or misunderstanding the detection limit calculation or decimals/measurement units are not correctly given.

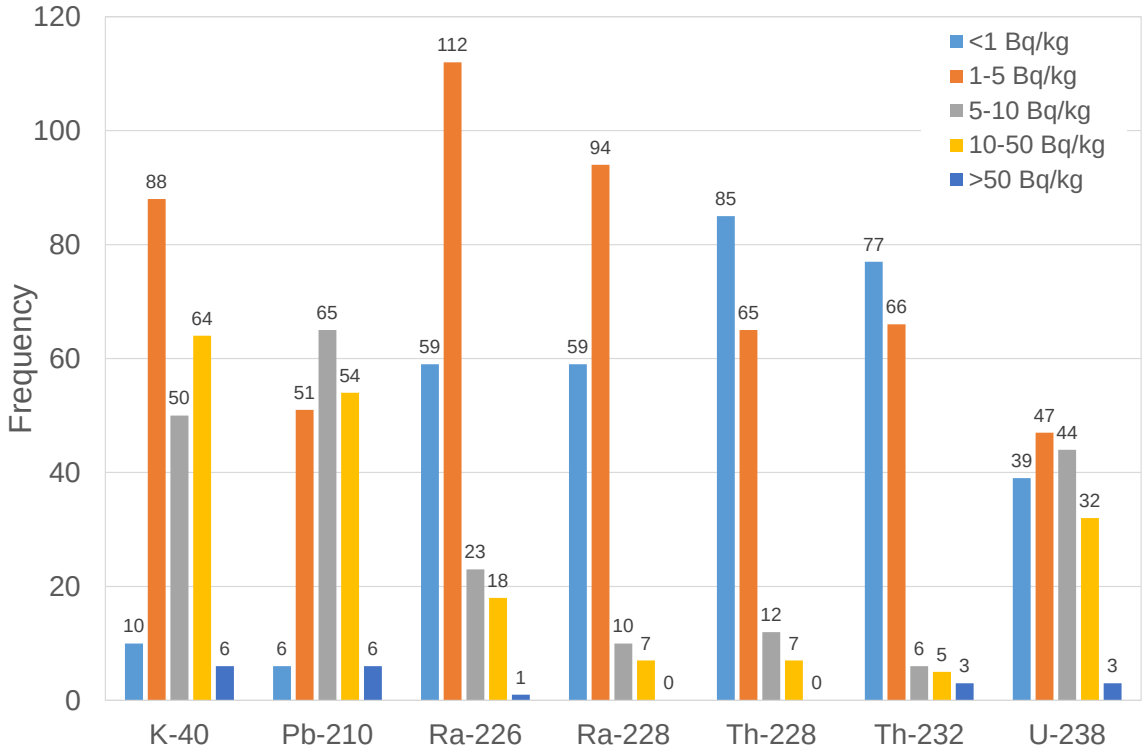
Another observation was that the majority of participants did not indicate measurement units. When doing so it was in different forms: Bq/kg, mBq/kg, Bq/kg dry mass or ppm.

Table 1. Summary of reported detection limits for radionuclides and materials (Bq/kg dry mass).

Radionuclide	**NORM-01**	**NORM-02**	**NORM-03**
K-40	Median: 6.0 Bq/kg Reported range: 0-188 Bq/kg	Median: 6.1 Bq/kg Reported range: 0.1-336 Bq/kg	Median: 5.0 Bq/kg Reported range: 0.1-347 Bq/kg
Pb-210	Median: 8.2 Bq/kg Reported range: 0-109.4 Bq/kg	Median: 6.7 Bq/kg Reported range: 0-98.1 Bq/kg	Median: 6.2 Bq/kg Reported range: 0.26-100.8 Bq/kg
Ra-226	Median: 2.1 Bq/kg Reported range: 0-80.4 Bq/kg	Median: 2.0 Bq/kg Reported range: 0-32.4 Bq/kg	Median: 2.0 Bq/kg Reported range: 0-32.1 Bq/kg
Ra-228	Median: 1.7 Bq/kg Reported range: 0-29.3 Bq/kg	Median: 1.5 Bq/kg Reported range: 0-15.2 Bq/kg	Median: 1.3 Bq/kg Reported range: 0-12.0 Bq/kg
Th-228	Median: 1.1 Bq/kg Reported range: 0-34.2 Bq/kg	Median: 1.0 Bq/kg Reported range: 0-30.7 Bq/kg	Median: 0.9 Bq/kg Reported range: 0-27.0 Bq/kg
Th-232*	Median: 1.2 Bq/kg Reported range: 0-336.9 Bq/kg	Median: 1.0 Bq/kg Reported range: 0-302.2 Bq/kg	Median: 1.0 Bq/kg Reported range: 0-364.5 Bq/kg
U-238	Median: 6.0 Bq/kg Reported range: 0-80.0 Bq/kg	Median: 4.6 Bq/kg Reported range: 0-56.0 Bq/kg	Median: 4.4 Bq/kg Reported range: 0-70.0 Bq/kg

**Note that Th-232 is not measured by gamma-ray spectrometry so the limits reported for gamma-ray spectrometry are generally from measurement of Ra-228 via its daughter Ac-228. This table gives an overview of limits considering all techniques.*
Source: JRC

Figure 19. Histogram of reported detection limits for the radionuclides of interest.



Source: JRC

As seen from **Figure 19**, the majority of detection limits are below 10 Bq/kg for all seven radionuclides. However, the frequency of higher detection limits increased for K-40, Pb-210 and U-238 as compared to the other radionuclides.

Which method did you use for the calculation of the detection limits?

— ISO 11929: 38 participants

— Currie's method: 39 participants

— Other: 8 participants
1) 3 times the error in the origin of the calibration line; 2) Traditional ORTEC method by GammaVision (ORTEC); 3) work procedure; 4) MAR(Bq)=4.66 (radq(counts))/(livetime×eff×I); 5) internal method of equipment producer; 6) KTA algorithm; 7) EPA 6020B; 8) gamma-ray spectrometry.

3.2.2.4 Uncertainty budget and its components with relative uncertainty (%)

Regarding uncertainty, for alpha-particle spectrometry and gamma-ray spectrometry 9 and 80 replies were submitted, respectively. Whereas for ICP-MS and LSC, one reply was submitted for each detection technique. The reported uncertainty components for the different measurement methods are summarised in **Table 2**.

Table 2. Typical uncertainty components of the measurement techniques (as given by the participants).

Technique	**Uncertainty components and relative uncertainty (%)**	**Number of replies**
alpha-particle spectrometry	counting statistics of tracer and radionuclide of interest (1-20%), efficiency calibration (0.1-5%), chemical yield (1-5%), background (0.5-15%), repeatability (25%), weighing (0.05-1%), activity concentration of tracers (2%), analyst, sample inhomogeneity (0.5%)	9
gamma-ray spectrometry	Counting statistics (0.05-30%), efficiency calibration (0.04-30%), efficiency transfer/peak fitting (1-12%), geometry-filling height (0.05-7%), weighing (0.05-5%), background (0.5-28%), moisture determination (0.001-4.5%), Certified Reference Material-reference source (2-3%), nuclear data library (e.g. emission probability, cascade correction, half-life/decay constant) (<0.1-1.7%), attenuation correction (0.3-10%), Repeatability(2-7%), true coincidence summing correction (1.5-5%), self- absorption correction factor (1-5%), chemical composition and density (0.04-2%), position of sample (<0.1-2%), difference in sample and calibration source geometry (0.5%), sample inhomogeneity (5%), oscillations of the instrumental response (1.4%), volume (0.5%), analyst, dead-time+others (5%)	80
ICP-MS	Repeatability (12%), calibration (6%), recovery (10%)	1
liquid scintillation counting	Mass (0.1%), efficiency calibration (0.1%), repeatability (25%), chemical yield (2%), Poisson statistics	1

Source: JRC.

There was a comment from a laboratory using gamma-ray spectrometry, that "*Uncertainty budgets different for each sample and nuclide so can't provide in general form.*"

As seen, the participants identified more components (n>20) for the gamma-ray spectrometry uncertainty budget than for any other measurement techniques. This is also due to that gamma-ray spectrometry was the most frequently used measurement technique in this proficiency test therefore, higher number of replies received from the participants on that.

The general observation is that participants assigned uncertainties to each measurement results for almost all measurement techniques. In case of gamma-ray spectrometry measurements there were few exceptions where results were submitted but their uncertainties were not given in 19 cases which corresponds to approximately 1.3% of the gamma-ray spectrometry results.

As reported by Hult et al. (2024a), out of the 1586 submitted results, 94.7% of the z scores and 73.1% of the zeta scores were acceptable. The overall performance seems satisfactory when looking at the z- scores. However, uncertainty budget still needs further scrutiny as close to 30% of the zeta scores were questionable or unacceptable. Therefore, the spread of uncertainties was checked and the median uncertainties were calculated from the participants submitted data and presented in Table 3- 6. In **Table 3** the overall uncertainties are presented, where all the reported data was pooled and then sorted as a function of measurement techniques only. The uncertainties for alpha-particle spectrometry measurements of Th-228, Th-232 and U-238 were also collected in

Table 4. Uncertainty data was further sorted according to measured radionuclides (**Table 5**) and PT material (**Table 6**) in case of gamma-ray spectrometry but not for the rest of the measurement techniques due to very limited data. Uncertainties were calculated as relative standard uncertainty with a coverage factor of 2. When a participant indicated other coverage factor, then their uncertainty was corrected accordingly.

Table 3. Overall uncertainties of the different measurement techniques.

Technique	**Median-, minimum and maximum relative expanded uncertainty values (at k=2 coverage factor)**
alpha-particle spectrometry	12.5% (2.5-54.5%)
gamma-ray spectrometry	12.3% (0.5-96.6%)
ICP-MS	26.6% (13.9-29.0%)
ICP-OES	0.8% (0.2-70.4%)
liquid scintillation counting	26.6% (25.0-49.9%)
Solid scintillation (ZnS(Ag))	11.6% (9.6-15.3%)

Source: JRC

In **Table 3** the highest data spread can be observed for gamma-ray spectrometry, ICP-OES and alpha-particle spectrometry. The smallest expanded uncertainties (even below 1%) were identified for gamma-ray spectrometry and ICP-OES. Considering the uncertainties from the different components in the uncertainty budget, these <1% uncertainties are not realistic for gamma-ray spectrometry.

Uncertainties of alpha-particle spectrometry for Th-228, Th-232 and U-238 are given in Table 4.

Table 4. Uncertainties of alpha-particle spectrometry for Th-228, Th-232 and U-238.

Radionuclide	**Median-, minimum and maximum relative expanded uncertainty values (at k=2 coverage factor)**
Th-228	12.5% (2.5-34.0%)
Th-232	13.3% (7.4-54.5%)
U-238	12.0% (2.7-21.4%)

Source: JRC

It can be noted that, the lowest uncertainties were obtained for U-238 and Th-228 measurements and in general for U-238 measurements realistic and smaller uncertainties were achieved than for the thorium measurements.

Uncertainties from gamma-ray spectrometry measurements were sorted as a function of radionuclides and materials in **Table 5** and **Table 6**.

Table 5. Uncertainties of gamma-ray spectrometry measurements for U-238, Ra-226, Pb-210, Th-232, Ra-228, Th-228 and K-40.

Radionuclide	**Median-, minimum and maximum relative expanded uncertainty values (at k=2 coverage factor)**
K-40	8.8% (0.9-29.1%)
Pb-210	25.4% (4.9-85.0%)
Ra-226	10.8% (0.5-42.2%)
Ra-228	10.5% (0.9-77.3%)
Th-228	11.2% (0.8-32.1%)
Th-232	11.0% (0.6-53.0%)
U-238	20.0% (1.1-96.6%)

Source: JRC

Considering the median uncertainties, the order from lowest to highest values is the following: K-40, Ra-228, Ra-226, Th-232, Th-228, U-238 and Pb-210. The spread of uncertainties follows almost the same order, where K-40, Th-228, Ra-226 and Th- 232 varies the least while U-238 and Pb-210 show the highest variation followed by Ra-228.

Table 6. Uncertainties of gamma-ray spectrometry measurements linked to PT materials.

PT material	**Median-, minimum and maximum relative expanded uncertainty values (at k=2 coverage factor)**
NORM-01	11.6% (0.5-85.0%)
NORM-02	13.3% (0.6-96.6%)
NORM-03	12.5% (0.6-84.2%)

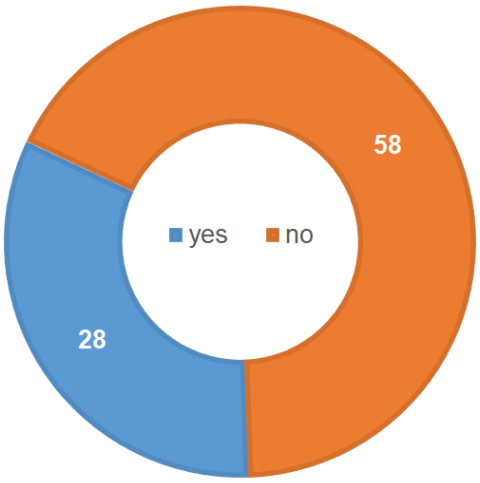
Source: JRC

The spread of uncertainty data was the largest for NORM-02 material and for the other two materials almost the same. Nevertheless, the median value of NORM-02 does not seem significantly different from that of the other two PT materials.

3.2.2.5 Did you encounter any difficulties while processing or measuring any of the PT reference materials?

The number of laboratories experiencing difficulties in measuring PT reference materials are presented in **Figure 20**.

Figure 20. Number of laboratories experiencing any difficulties while measuring PT reference materials.



Source: JRC

Description of difficulties

- crushing and measuring the sample NORM-2: 20 participants
- Laboratory activity was interrupted due to COVID cases: 2 participants
- the chemical composition of samples was missing: 2 participants
- insufficient time for analysis
- lack of tools to process block type materials
- insufficient amount of material to fully-fill sample container
- could not seal the geometry
- Digestion process took longer
- Increased counting time to achieve good counting statistics
- Dust and contamination of laboratory due to processing NORM 2
- Samples were sent in different form than requested by the participant (asked for a max diameter of 1 cm), had to buy tools

4 Participants' feedback on the REM 2020 PT exercise

In the reporting survey, participants had the opportunity to give feedback and make comments about the proficiency test let it be technical or organisational related aspects. The overwhelming majority of comments were positive. The participants were grateful for the opportunity to participate in this PT. For some, it is a unique occasion to participate in a PT due to costs. This kind of thematic PT with the coordination of an impartial and independent organisation is not common. In the analysis, the feedback submitted by the participants were categorised as "Comments and remarks", "Compliments" and "Benefits of this PT" and presented in the next sections.

4.1 Comments and remarks

- The samples were differently named (e.g.: Norm 1, cement), see also the reporting website and the reporting survey.
- The questionnaire should be more descriptive, allowing to enter information for any different geometries and tools used for analyzing the samples.
- a more flexible questionnaire to better explain the different analytical geometries used for each sample
- Difficult to measure NORM-2 type samples
- Challenging exercise with some non-trivial radionuclides.
- not enough of sample material for full filing of container
- would be useful to discuss different methods for the determination of the absorption coefficient
- In the case of block sample (NORM 2), there was a concern about sample inhomogeneity during in-house treatment.
- unlucky planning, coincidence with Covid-19 pandemia.
- When Monte Carlo simulation is used for detector efficiency and efficiency correction factors, the chemical composition would be desirable
- Due to the busy schedule of the lab, only ICP-OES analysis was done

4.2 Compliments

- Good/well/excellent/interesting/OK/useful proficiency test: 18 participants
- Clear instructions, swift communication
- Laboratories would like to participate in future JRC proficiency tests

4.3 Benefits of this PT

- The PT allows i) the validation of analytical methods and quality control internal routines and ii) to detect unexpected problems in the analytical work.
- This PT provided useful reference NORM materials that can be used for future measurements.

5 Future proficiency tests

In the reporting survey, PT participants could also indicate which radionuclides and matrices would they consider useful in the future proficiency tests. From the 85 participants who filled in the reporting survey, 39 replied for this question and their "wish-list" is presented in **Table 7** below. It was possible to mention more than one matrix and radionuclides. Additional comments were also given and introduced in the last column. The reason for blank cells in the table is that sometimes participants did not give both information (matrix and radionuclides) but only one of them.

Table 7. List of radionuclides and matrices for future proficiency tests.

Matrices (number of mentions)	**Radionuclides/Parameters**	**Comments from participants**
NORMs (13): phosphogypsum, clinkers, ashes, high density slags, zeolites, residues from metallic mining processes	Same radionuclides as REM2020 PT, Th-230, Po-210 Short-lived nuclides, radionuclides not in equilibrium	
Water (11)	Rn-222, Ra-226 and Ra-228 gross alpha/beta activity natural and artificial gamma emitters in water	
Air/Aerosol filter (5)	Pb-210 and Po-210	
Soil (5)	Cs-137, Natural and anthropogenic radionuclides	
Food/Feed (3)	Cs isotopes, transuranic elements, radium, Pb-210 and Sr-89/90	
Building materials (3) (concrete, cements)	Same radionuclides as REM2020 PT	Cements: "mixed with NORM material from a circular economy or after recycling"
Animal bones (1)	Pu, Am, Sr	
Resins and metal shavings (1)	Natural and artificial radionuclides	
Charcoal (1)	I-131	
Milk powder (1)	-	
Solid matrices (1) volcanic lava (1) marine sediments (1)	-	"high density materials containing heavy elements; to test numerical tools."
"Dry residues of water samples" (1)		
Powders (non-specified) (1)	-	
No matrix was given (2)	Po-210, "All natural radionuclides, Artificials"	

Source: JRC

The most frequently mentioned input was NORMs and the related natural radionuclides. The second most wanted matrix was water, especially radon-in-water or radium isotopes in water, followed by filters, soil and food/feed matrices. There were some less-common matrices mentioned (e.g. volcanic lava, resins) and some very generic, non-specific answers were also given on this question (e.g. “powders” or “natural and artificial radionuclides”). This can be also noted, that apart from the natural radionuclides, there is an increased interest in including artificial/transuranic radionuclides too.

6 Follow-up workshop

A hybrid-workshop (in-person and on-line) dedicated to the REM2020 proficiency test was organised by the JRC-Geel, Radionuclide Metrology team between 24-26 September 2024. The objective of this workshop was to discuss predominantly the NORM-construction material measurement related issues with the aim to improve the level of radiation monitoring in Europe to ensure the health and safety of citizens. The details and outcome of the workshop are presented in a report (Hult and Paepen, 2024b).

There were 88 registered participants from 28 countries, representing environmental radioactivity monitoring laboratories, metrology institutes, universities, regulatory bodies, Euratom Article 35-36 experts, experts from other JRC directorates and Directorate General for Energy (DG ENER). During the three-day event, presentations were given by invited speakers and also participants had the opportunity to present their work on linked to the following topics:

- REM2020 building material PT
- Reference materials production, PT reference value assignment
- Gamma-ray spectrometry and alpha-particle spectrometry measurements of NORMs
- A new European standard on measuring radioactivity in building material
- Other scientific disciplines (geology, hydrogeology) linked to NORMs
- NORMs and construction materials
- Nuclear decommissioning
- Communication of risks linked to ionising radiation

7 Additional discussions on measurement method performances

On top of the analysis of the on-line survey, the spreadsheets containing information about the measurement methods, submitted measurement results and their uncertainties were also briefly reviewed. The applied analytical methods, number of participants and reported measurement values are listed in **Table 8**.

Table 8. Applied analytical methods and number of reported measurement values.

Analytical methods	**Number of participants**	**Number of reported values**	**Radionuclides**	**Notes**
alpha-particle spectrometry	12	77	U-238, Th-232, Th-228	31, 33 and 13 values respectively per radionuclide
gamma-ray spectrometry	94	1449	All seven (Th-232 indirectly)	
ICP-MS	2	10	U-238, Th-232	One lab all three materials, another lab NORM-01 and NORM-03
ICP-OES	1	7	U-238, Th-232, K-40	all three materials
LSC	1	6	Ra-226, Pb-210	all three materials
ZnS(Ag) solid scintillation after co-precipitation	1	3	Ra-226	all three materials
Not reported		32	-	-
Total	96	1586	All seven	-

Source: Hult et al., 2024a and JRC

It is seen and as expected, gamma-ray spectrometry was the most used method since the majority of the requested radionuclides (U-238, Ra-226, Pb-210, Th-232, Ra-228, Th-228 and K-40) in construction materials can be measured directly with this technique without applying destructive sample preparation methods. Almost every participant (97.9% of them), who reported measurement results, used gamma-ray spectrometry submitting 91% of the total number of results. However, in certain cases spectral interferences can make gamma-spectrometry measurements difficult. For example, using the 186.2 keV line for Ra-226 determination due to the

interference from the 185.7 keV line from U-235. Moreover, in case of Ra-226 determination extra time is needed to reach equilibrium between Ra-226, Rn-222 and their short-lived decay products in the decay chain. This requires a 3-4 weeks of storage time until measurements. Th-232 was determined either as mean of Ra-228 and ^{228}Th or taken as equal to that of Ac-228.

Alpha-particle spectrometry was the second most used method where 12 participants (12.5% of the participants) submitted 77 results (4.9% of the total results). Alpha-particle spectrometry needs destructive sample preparation in case of construction material analysis. The procedure should be able to perform total dissolution of material to be able to recover all fraction from the radionuclides of interest. In the environmental radioactivity monitoring laboratories alpha-particle spectrometry is a routine method for determining alpha-particle emitting radionuclides, in this case U-238, Th-232 and Th-228. This method could be also suitable for Ra-226 determination but requires labour intense sample preparation and the alpha source preparation based on $BaSO_4$ co-precipitation is also a manipulation that requires proficient analysts. What might also compromise the accuracy of the alpha spectrometry approach is lack of available long-lived radium tracer. The most common tracer for Ra-226 analysis is Ba-133 due to their chemical similarities. However, there are still some differences between the behaviour of radium and barium in certain chemical reactions (e.g. precipitation, ion exchange) that might result in different Ra-226 and Ba-133 recoveries (Sill, 1987/Lozano et al., 1997).

ICP based techniques, especially ICP-MS, are considered state of the art techniques but investing in such instruments is still very expensive. The biggest advantages are that they are very accurate and rapid techniques, analytical results can be achieved within an hour not only about one but multi-elements (including radionuclides). However, the analysts have to be sure that solid materials are totally dissolved and no residues are left resulting in loss of analyte of interest. Furthermore, spectral interferences should be taken care of and corrected for (see isobar interferences and molecule-ion formation). The incomplete sample digestion and spectral interferences could be also accountable for the non-satisfactory performance of ICP-MS technique (e.g. Th-232 measurement in NORM-02 sample). Additional possible error could be the incorrect conversion of results from massic unit to activity unit.

LSC and solid scintillation methods were also underrepresented in this PT probably due to extensive sample preparation and potential interferences in the measurements. On the other hand, they are accurate techniques when less complex samples are measured (e.g. widely used in water analytics).

7.1 Recovery

For destructive analytical methods, it worth analysing recoveries of radionuclides of interests. Recovery is a method characteristic that can indicate how efficient a method is and can also provide information about robustness. Recoveries of K-40, Pb-210, Ra-226, Th-228, Th-232 and U-238 obtained with various destructive methods are presented in **Table 9**.

Table 9. Recovery of radionuclides from the applied destructive analytical methods.

Methods	**NORM-01**	**NORM-02**	**NORM-03**
Alpha-particle spectrometry	Th-228: 63% (7.6-100%) Th-232: 51% (7.6-100%) U-238: 62% (19.2-100%)	Th-228: 69% (6.6-100%) Th-232: 61% (6.6-100%) U-238: 63% (17-100%)	Th-228: 75% (22-100%) Th-232: 72% (22-104%) U-238: 70% (39-100%)
ICP-MS	Th-232: 100-120% U-238: 81-100%	Th-232: 120% U-238: 81%	Th-232: 100-120% U-238: 81%-100%
ICP-OES	Th-232, U-238, K-40: 55%	Th-232, U-238, K-40: 55%	Th-232, U-238, K-40: 55%
Liquid scintillation counting	Ra-226, Pb-210: 100%	Ra-226, Pb-210: 100%	Ra-226, Pb-210: 100%
ZnS(Ag) solid scintillation	Ra-226: 100%	Ra-226: 91.1%	Ra-226: 81.5%

Source: JRC

Mean values are given only for alpha-particle spectrometry because of the limited reported measurement data from other analytical techniques.

For all three radionuclides, the mean recoveries seem to be slightly improved from NORM-01 to NORM-03 as seen in Table 10. However, when the individual recovery data are scrutinised, then similar recovery success rates can be observed for all three NORM materials and radionuclides as well. The recovery of >50% for Th-228 was for 9 out of 13 measurements; for Th-232, 21 out of 33 measurements achieved >50% recovery. While in case of U- 238, 20 out of 28 measurements achieved >50% recovery which is slightly better ratio than for the other radionuclides. Despite the complex matrices, alpha spectrometry proved to be very robust method with consistent performance.

7.2 Changing standard deviations for proficiency assessment (σ_{pt})

Regarding accuracy, a simulation was done to see how the z scores would change when more stringent standard deviations for proficiency assessment (σ_{pt}) would be applied (**Table 10**). Two cases were introduced. In *Case 1* we decreased the σ_{pt} from 20% to 15% and in *Case 2* to 7.5% for K-40, Ra-226, Ra-228, Th-228 and Th-232. While for U-238 and Pb- 210 σ_{pt} was decreased from 30% to 20% (*Case 1*) and then to 10% (*Case 2*) for all three NORM materials.

Table 10. Overview on participants' data with statistics on the number of reported results with initial z scores and z-scores after changing the standard deviations for proficiency assessment (σ_{pt}).

Sample	Radio-nuclide	No. of reported values	Initial sigma PTs						Case 1						Case 2					
			z score: No. of submitted results			z score: % of submitted results			z score: No. of submitted results			z score: % of submitted results			z score: No. of submitted results			z score: % of submitted results		
			|z| ≤ 2	2<|z|<3	|z| ≥ 3	|z| ≤ 2	2<|z|<3	|z| ≥ 3	|z| ≤ 2	2<|z|<3	|z| ≥ 3	|z| ≤ 2	2<|z|<3	|z| ≥ 3	|z| ≤ 2	2<|z|<3	|z| ≥ 3	|z| ≤ 2	2<|z|<3	|z| ≥ 3
NORM1 - Cement	K-40	94	92	1	1	97.9	1.1	1.1	90	2	2	95.7	2.1	2.1	72	15	7	76.6	16.0	7.4
NORM1 - Cement	Pb-210	75	66	4	5	88.0	5.3	6.7	57	5	13	76.0	6.7	17.3	35	13	27	46.7	17.3	36.0
NORM1 - Cement	Ra-226	91	87	1	3	95.6	1.1	3.3	85	2	4	93.4	2.2	4.4	74	10	7	81.3	11.0	7.7
NORM1 - Cement	Ra-228	77	74	2	1	96.1	2.6	1.3	73	2	2	94.8	2.6	2.6	67	4	6	87.0	5.2	7.8
NORM1 - Cement	Th-228	72	68	3	1	94.4	4.2	1.4	66	3	3	91.7	4.2	4.2	62	3	7	86.1	4.2	9.7
NORM1 - Cement	Th-232	75	71	1	3	94.7	1.3	4.0	66	2	7	88.0	2.7	9.3	63	2	10	84.0	2.7	13.3
NORM1 - Cement	U-238	76	73	0	3	96.1	0.0	3.9	70	3	3	92.1	3.9	3.9	56	10	10	73.7	13.2	13.2
NORM2 - Expanded clay block (piece)	K-40	77	74	2	1	96.1	2.6	1.3	72	3	2	93.5	3.9	2.6	60	9	8	77.9	11.7	10.4
NORM2 - Expanded clay block (piece)	Pb-210	64	60	3	1	93.8	4.7	1.6	55	3	6	85.9	4.7	9.4	42	10	12	65.6	15.6	18.8
NORM2 - Expanded clay block (piece)	Ra-226	75	68	2	5	90.7	2.7	6.7	62	6	7	82.7	8.0	9.3	51	5	19	68.0	6.7	25.3
NORM2 - Expanded clay block (piece)	Ra-228	65	64	0	1	98.5	0.0	1.5	62	1	2	95.4	1.5	3.1	52	2	11	80.0	3.1	16.9
NORM2 - Expanded clay block (piece)	Th-228	64	60	0	4	93.8	0.0	6.3	57	2	5	89.1	3.1	7.8	45	10	9	70.3	15.6	14.1
NORM2 - Expanded clay block (piece)	Th-232	60	54	2	4	90.0	3.3	6.7	51	2	7	85.0	3.3	11.7	42	6	12	70.0	10.0	20.0
NORM2 - Expanded clay block (piece)	U-238	66	62	1	3	93.9	1.5	4.5	58	2	6	87.9	3.0	9.1	50	8	8	75.8	12.1	12.1
NORM3 - Expanded clay block (powder)	K-40	93	93	0	0	100.0	0.0	0.0	91	2	0	97.8	2.2	0.0	80	8	5	86.0	8.6	5.4
NORM3 - Expanded clay block (powder)	Pb-210	75	70	2	3	93.3	2.7	4.0	59	9	7	78.7	12.0	9.3	41	14	20	54.7	18.7	26.7
NORM3 - Expanded clay block (powder)	Ra-226	90	84	2	4	93.3	2.2	4.4	83	2	5	92.2	2.2	5.6	70	6	14	77.8	6.7	15.6
NORM3 - Expanded clay block (powder)	Ra-228	77	74	1	2	96.1	1.3	2.6	74	0	3	96.1	0.0	3.9	68	3	6	88.3	3.9	7.8
NORM3 - Expanded clay block (powder)	Th-228	72	68	0	4	94.4	0.0	5.6	67	1	4	93.1	1.4	5.6	60	6	6	83.3	8.3	8.3
NORM3 - Expanded clay block (powder)	Th-232	73	67	0	6	91.8	0.0	8.2	65	0	8	89.0	0.0	11.0	61	1	11	83.6	1.4	15.1
NORM3 - Expanded clay block (powder)	U-238	75	73	0	2	97.3	0.0	2.7	67	4	4	89.3	5.3	5.3	53	10	12	70.7	13.3	16.0
Sum		**1586**	**1502**	**27**	**57**				**1430**	**56**	**100**				**1204**	**155**	**227**			
Average						**94.7**	**1.7**	**3.6**				**90.2**	**3.5**	**6.3**				**75.9**	**9.8**	**14.3**

Source: JRC

Case 1: σ_{pt} =15% for K-40, Ra-226, Ra-228, Th-228 and Th-232, σ_{pt} =20% for U-238 and Pb-210.
Case 2: σ_{pt} = 7.5% for K-40, Ra-226, Ra-228, Th-228 and Th-232, σ_{pt} = 10% for U-238 and Pb-210.

As expected, more data becomes questionable and unsatisfactory after changing the standard deviations for proficiency assessment to stricter values but still, in Case 1, the overall scores are not much different from the initial scores (**Table 10**) only with some exceptions. However, in Case 2 deterioration of scores can be observed especially for Pb-210 and for Ra-226 and thorium isotopes too. This means on one hand, that the standard deviations for proficiency assessment applied in Case 2 would be too penalising for the participants, while on the other hand standard deviations for proficiency assessment used in Case 1 could be considered in the next PTs for all radionuclides and similar matrices.

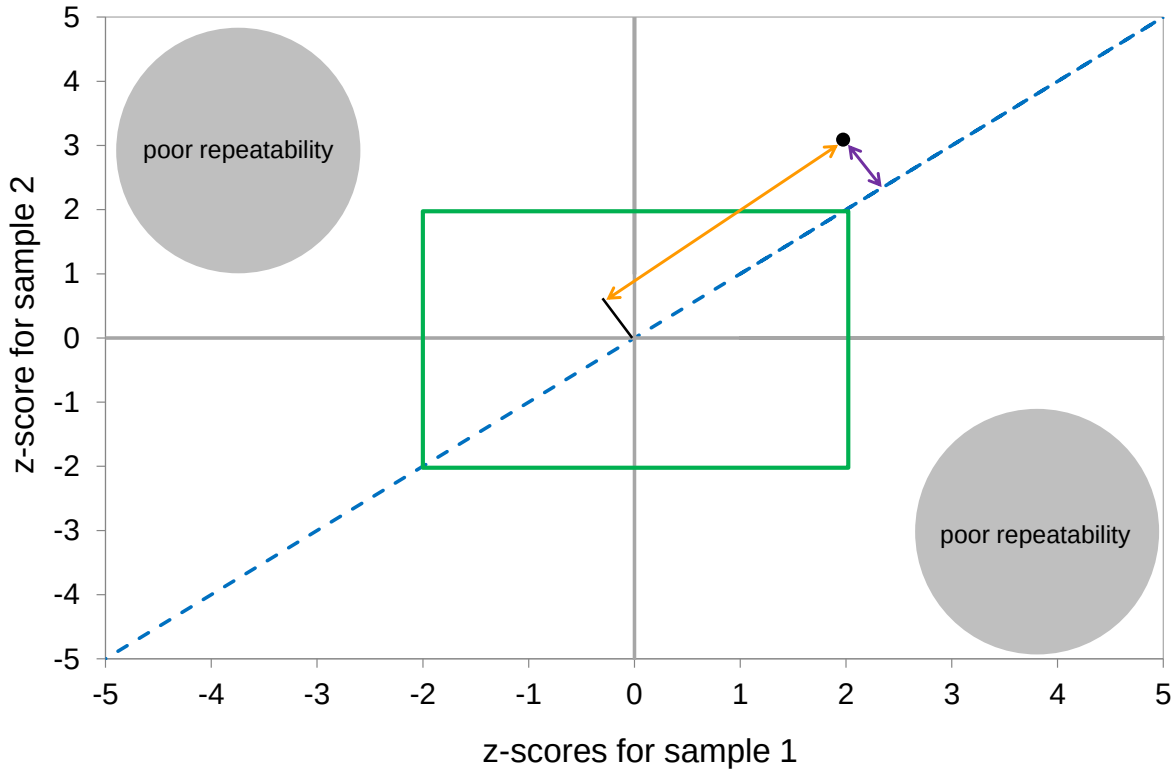
7.3 Youden plots

For measurement results obtained from the analysis of two similar proficiency test samples, Youden Plots can be used as a graphical approach to give information on repeatability and identify random/systematic errors as described by Youden (1959) and ISO 13528 (2015). A scatter plot is drawn in which the x-axis shows a performance evaluation score (e.g. z-scores) of sample 1 and the y-axis shows the same type of score for sample 2.

Youden Plots (see **Figure 21**) could be interpreted as following. The results can be grouped in four quadrants. When the variation in results is dominated by random errors, then the points are randomly distributed in all quadrants with approximately the same number of results in each quadrant. When systematic errors are significantly larger than random errors, then the points occur primarily in the upper right and lower left quadrants.

The distance of a point from the 45° line (blue dashed line) is proportional to the contribution of random error on the corresponding laboratory's results (purple arrow). The distance of a point from the zero points (intersection of the axes) on the 45° line is proportional to the laboratory's systematic error (orange arrow). Points in the far upper left and lower right quadrants show poor repeatability (grey circles). The acceptable results are distributed within the green box ($|z\text{-score}| \leq 2$).

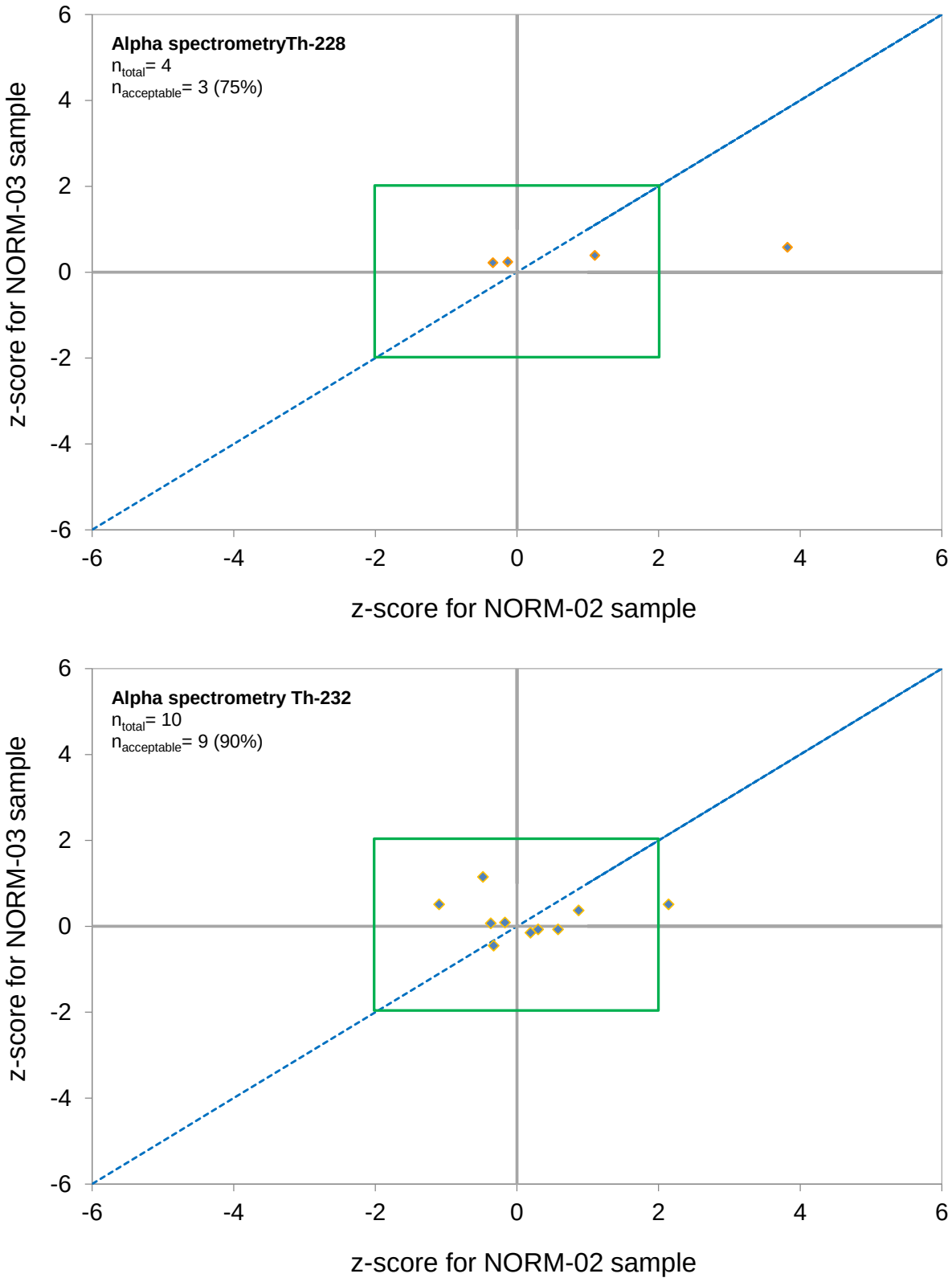
Figure 21. Interpretation of Youden Plot using z-scores. Green box: acceptable results (|z-score| ≤ 2). Purple arrow: the distance from blue dashed line contribution of random error on the laboratory's result. Orange arrow: the distance from the zero points is proportional to the laboratory's systematic error. Grey circles: poor repeatability.

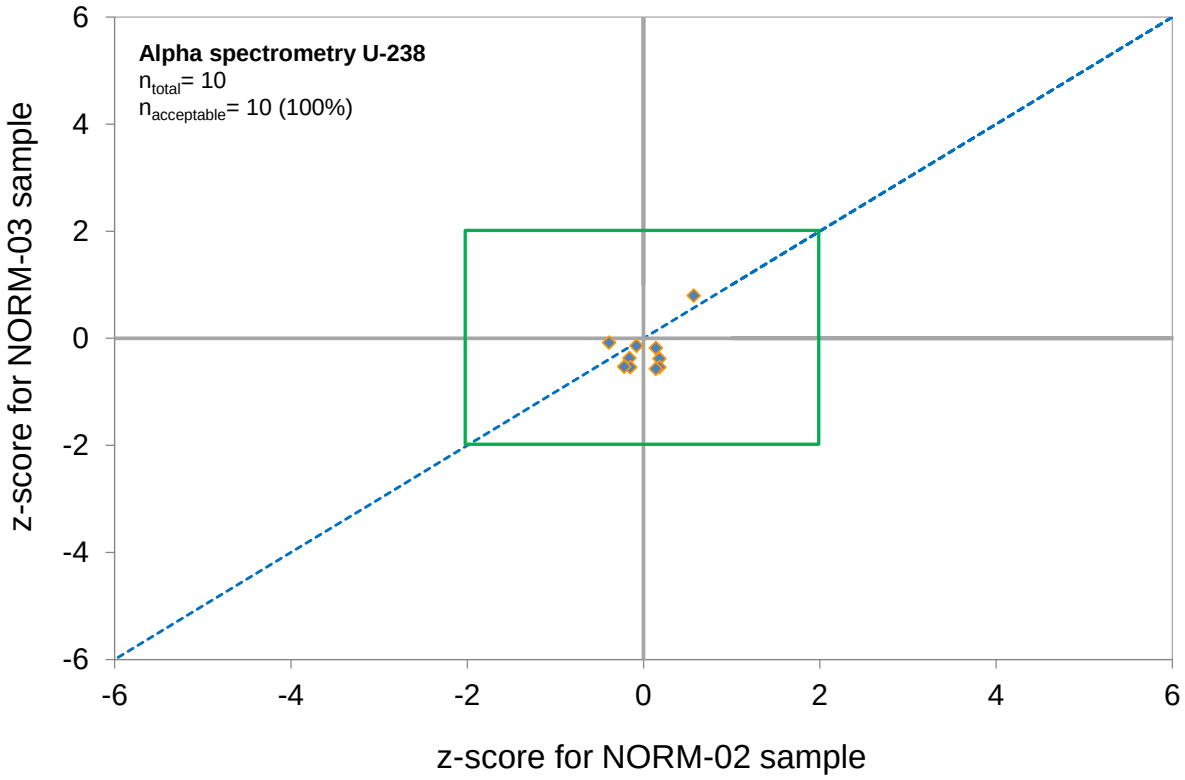


Source: JRC

Youden-plots were created for alpha-particle spectrometry (**Figure 22**) and gamma-ray spectrometry separately (**Figure 23**). In Youden-plots pairs of z-score values from the same type of PT sample but in different physical forms were used (NORM-02 and NORM-03). Every plot contains information on the total number of pairs of scores (n_{total}) and the number of acceptable scores ($n_{acceptable}$) and the percentage of acceptable scores in parenthesis.

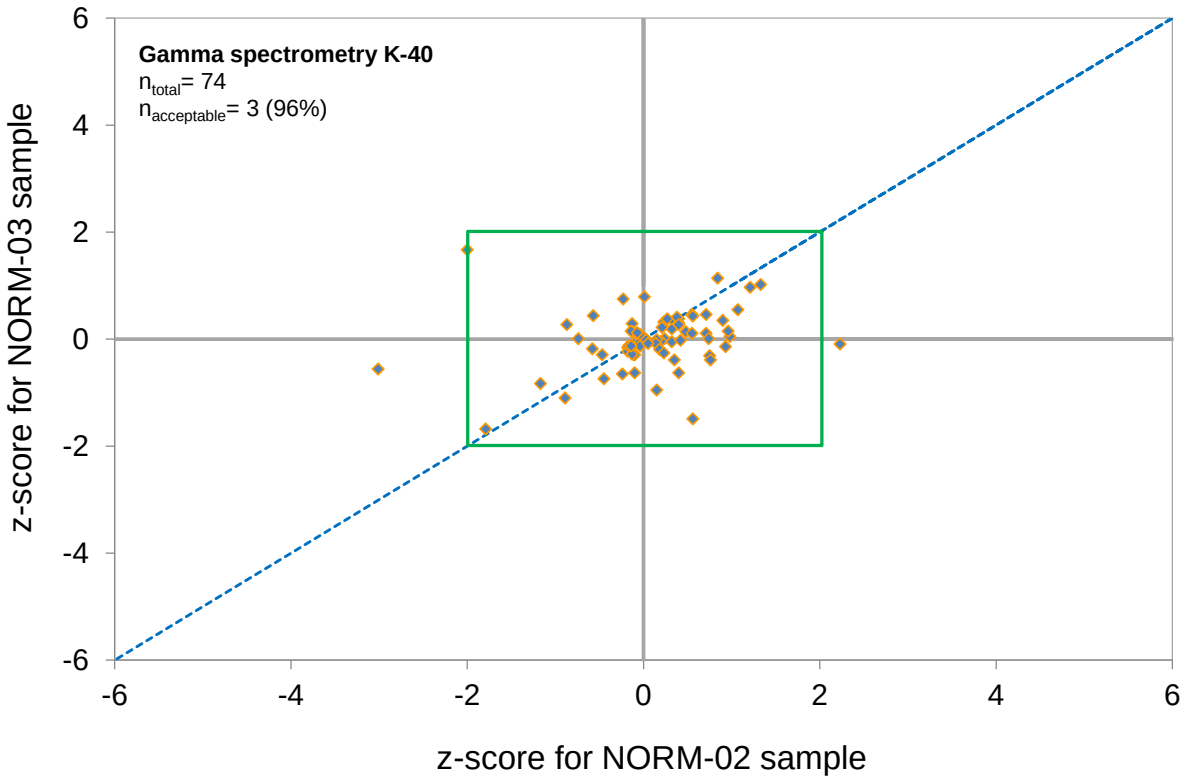
Figure 22. Three Youden Plots of z-scores for Th-228, Th-232 and U-238 alpha-particle spectrometry measurement results. The acceptable results are spread within the green box.

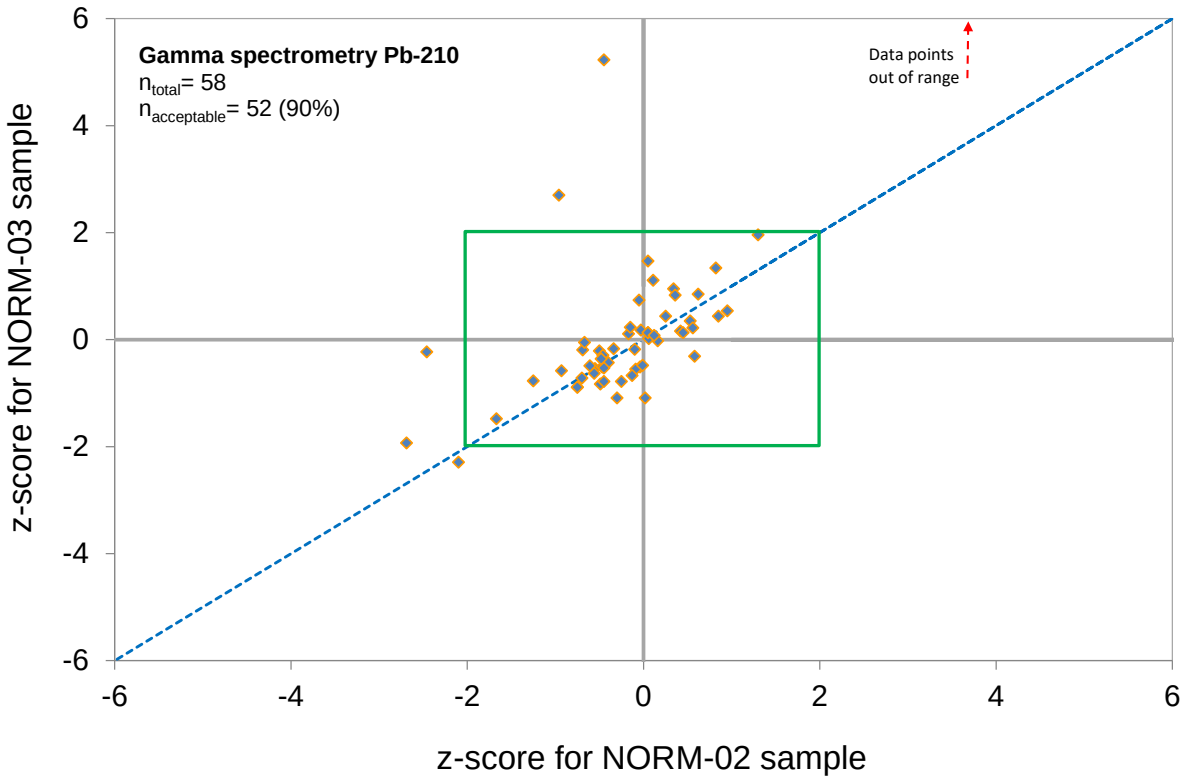


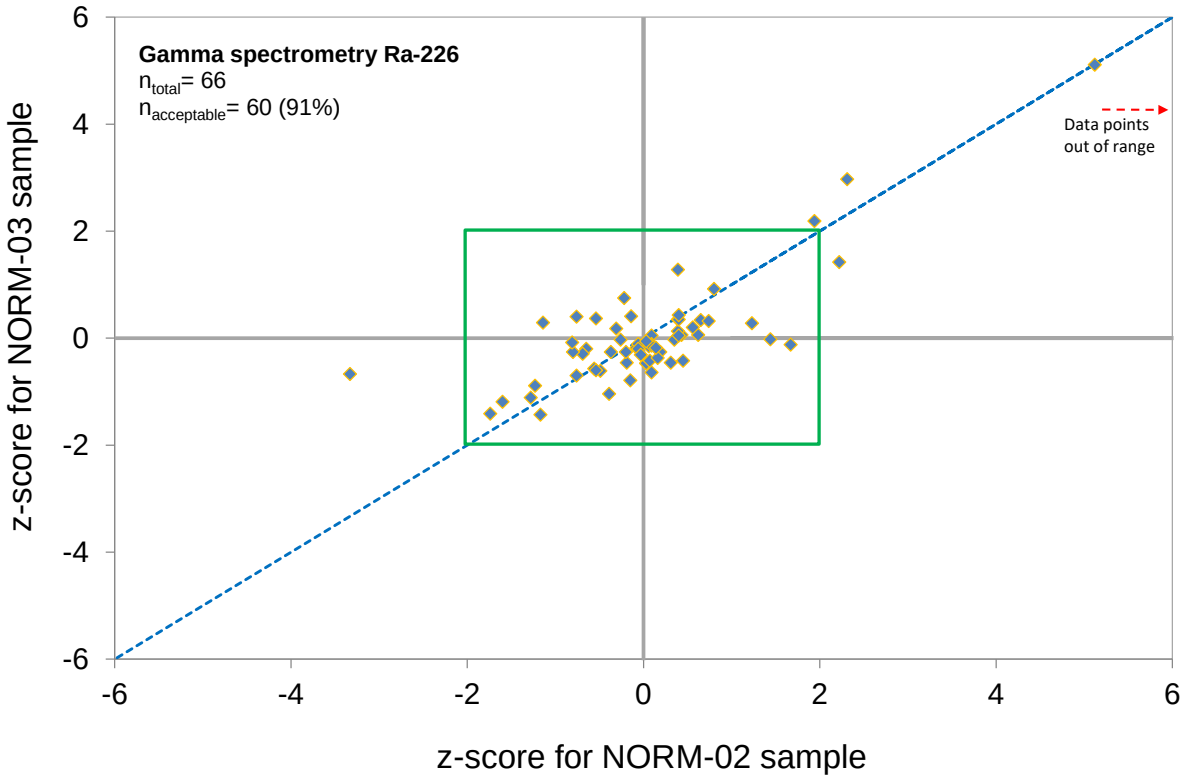

Alpha spectrometry U-238
n_total= 10
n_acceptable= 10 (100%)
z-score for NORM-03 sample
z-score for NORM-02 sample
6
4
2
0
-2
-4
-6
-6
-4
-2
0
2
4
6

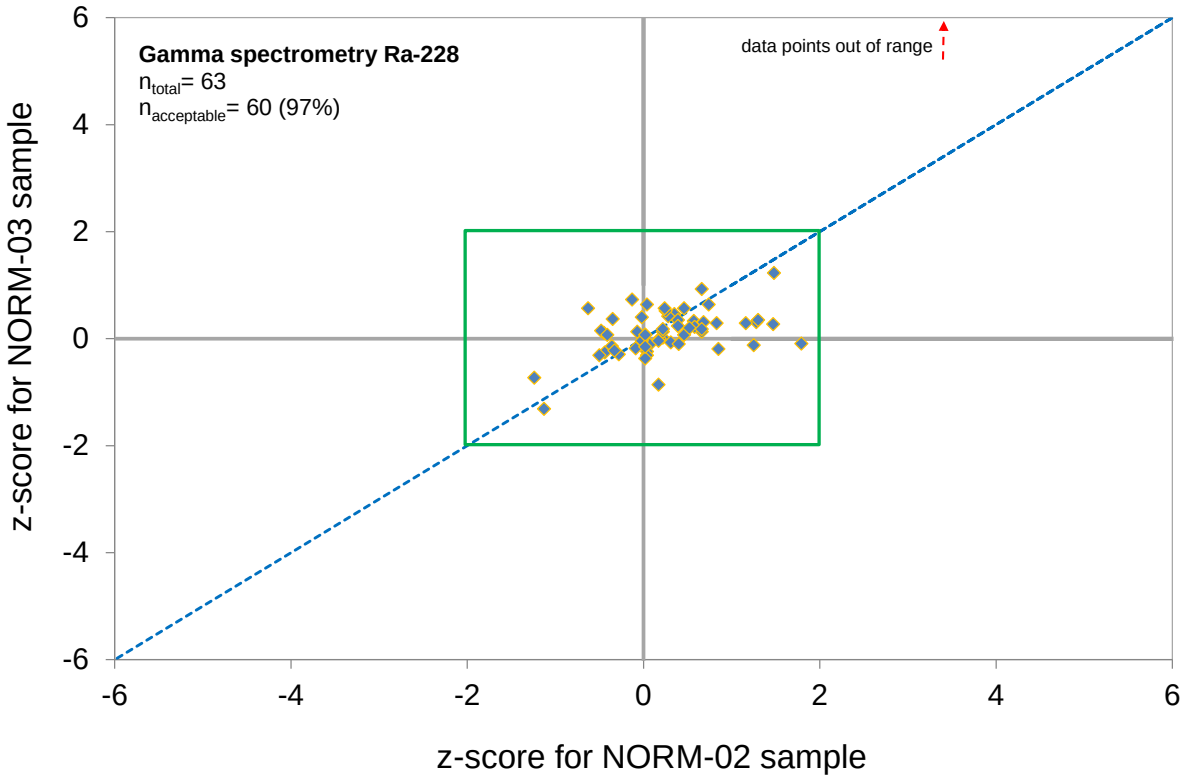
Source: JRC

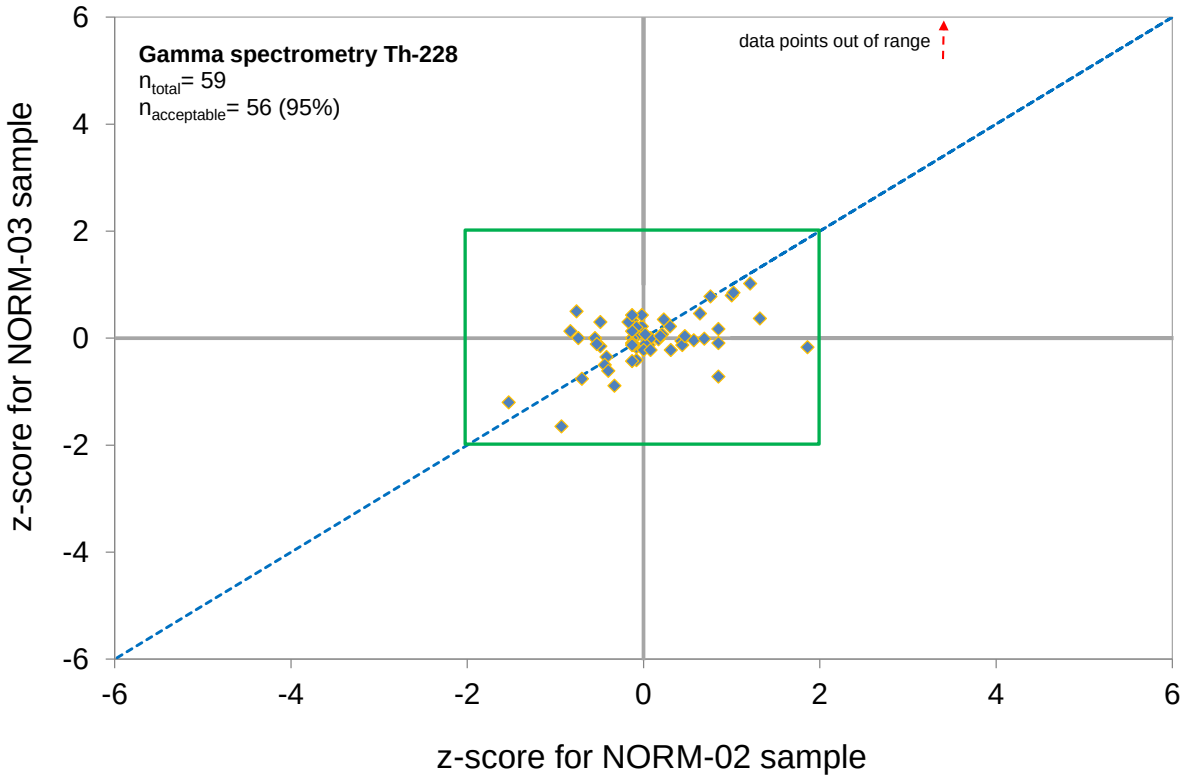
Figure 23. Seven Youden Plots of z-scores for all seven radionuclides from gamma-ray spectrometry measurement results. The acceptable results are spread within the green box.

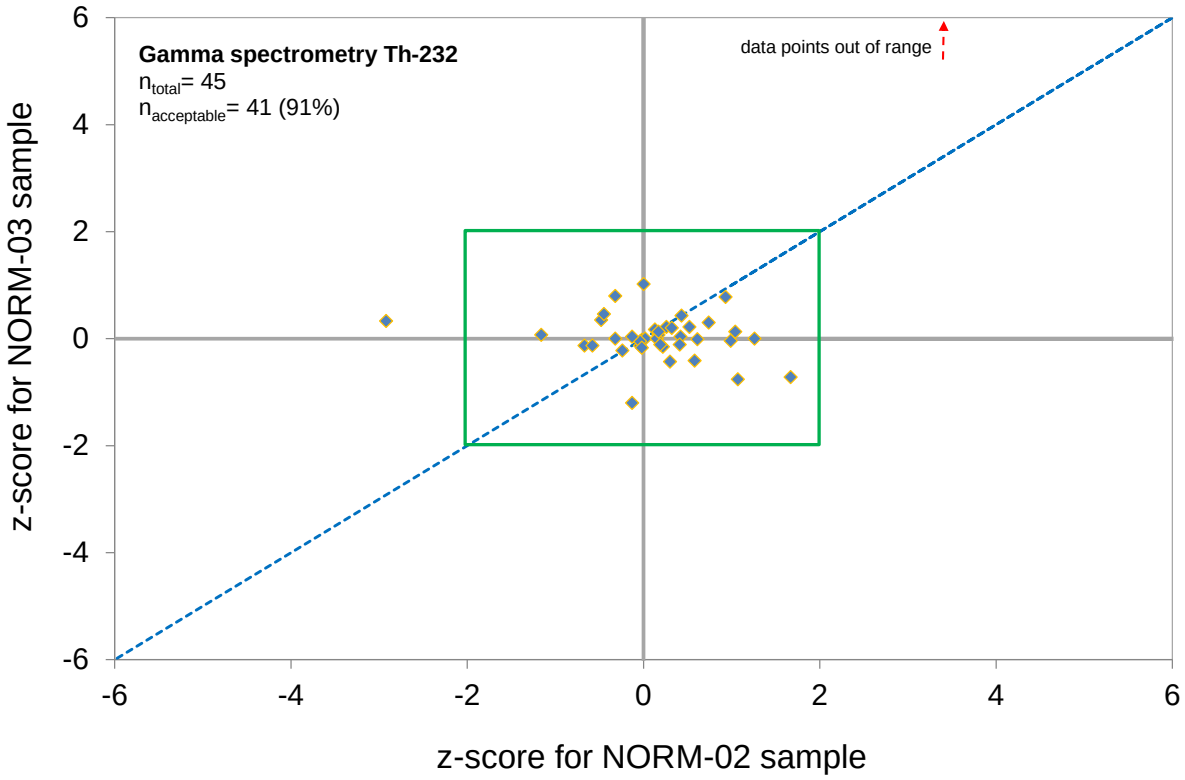


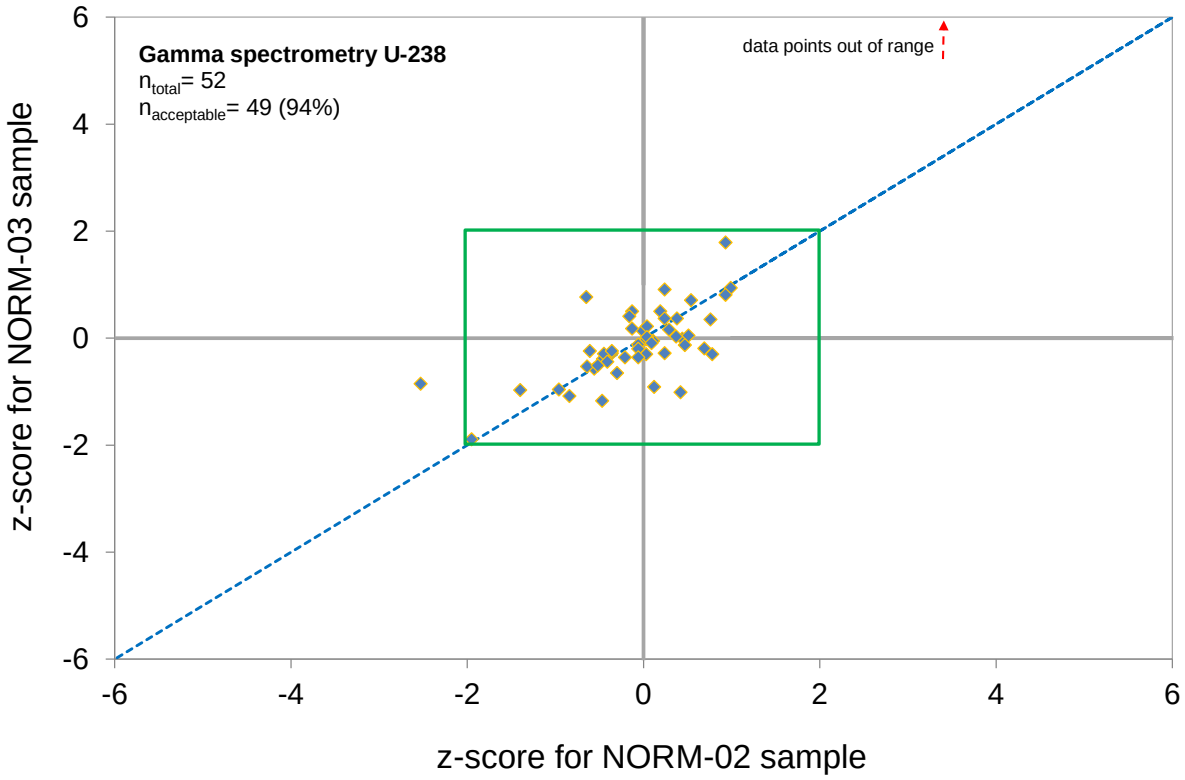



Gamma spectrometry Ra-226
n_total= 66
n_acceptable= 60 (91%)
Data points out of range
z-score for NORM-03 sample
z-score for NORM-02 sample
6
4
2
0
-2
-4
-6


Gamma spectrometry Ra-228
n_total= 63
n_acceptable= 60 (97%)
data points out of range
z-score for NORM-03 sample
z-score for NORM-02 sample
6
4
2
0
-2
-4
-6


Gamma spectrometry Th-228
n_total= 59
n_acceptable= 56 (95%)
data points out of range
z-score for NORM-03 sample
z-score for NORM-02 sample
6
4
2
0
-2
-4
-6


Gamma spectrometry Th-232
n_total= 45
n_acceptable= 41 (91%)
data points out of range
z-score for NORM-03 sample
z-score for NORM-02 sample
6
4
2
0
-2
-4
-6



Source: JRC

As seen in **Figure 22** and **Figure 23**, more than 90% of the measurement results for alpha-particle and gamma-ray spectrometry spread within the acceptable range, within one exception only (in case of Th-228 alpha spectrometry) where only four measurement pairs were submitted for the two different PT samples. On the basis of Youden plots, it can be concluded that both methods (alpha-particle and gamma-ray spectrometry) show good repeatability and accuracy for these types of samples even when the physical forms were different and the expanded clay block was difficult to process and homogenise. The Th-232 gamma-spectrometry results were based on its decay-products assuming secular equilibrium.

7.4 Method reproducibility

The international standard ISO 5725-2:2019, dealing with accuracy and precision, describes in details the determination of repeatability and reproducibility parameters for a standard method. The reproducibility standard deviation could be established through interlaboratory schemes where laboratories perform a number of independent measurements of the same sample under repeatability conditions.

In the REM2020 PT when high number of data is available for a measurement method (especially for gamma-ray spectrometry but also for alpha-particle spectrometry), calculation of method reproducibility could be done with an important condition. The same analytical methods were used in the different laboratories under repeatability conditions. Based on the data obtained from the participants about their methods it is not the case, since several different approaches were used (e.g. sample preparation, calibration etc.) within a measurement method category. However, the overall reproducibility was calculated for the gamma-ray spectrometry and for three radionuclide

measurements alpha particle spectrometry, with an ideal assumption that the same standardised methods were used (**Table 11**). The "*less-than values*" were not considered for this analysis.

Table 11. Overall reproducibility standard deviation of alpha-particle spectrometry and gamma-ray spectrometry for the three PT materials and radionuclides of interest. In brackets: Amount of data.

Alpha-particle spectrometry			
Radionuclide	**NORM-01**	**NORM-02**	**NORM-03**
Th-228	19.7% (n=5)	31.3% (n=4)	3.1% (n=4)
Th-232	13.6% (n=12)	17.4% (n=10)	8.3% (n=11)
U-238	10.3% (n=11)	8.1% (n=10)	13.4% (n=10)
Gamma-ray spectrometry			
Radionuclide	**NORM-01**	**NORM-02**	**NORM-03**
K-40	13.5% (n=93)	14.4% (n=75)	10.7% (n=92)
Pb-210	90.5% (n=69)	62.3% (n=59)	57.9% (n=69)
Ra-226	17.2% (n=89)	29.0% (n=72)	67.3% (n=88)
Ra-228	14.9% (n=76)	22.4% (n=63)	20.4% (n=76)
Th-228	36.1% (n=66)	78.2% (n=59)	48.5% (n=68)
Th-232	366% (n=55) 14.4% (n=54)*	462% (n=44) 22.8% (n=43)*	383% (n=55) 26.7% (n=54)*
U-238	33.9% (n=62)	134% (n=52) 20.9% (n=50)*	223% (n=61) 27.9% (n=60)*

Source: JRC

**without outliers*

Comparing the overall reproducibility of alpha spectrometry and gamma-ray spectrometry for the three commonly measured radionuclides (Th-228, Th-232, U-238), it can be observed that alpha-particle spectrometry exhibits better reproducibility than gamma-ray spectrometry. It has to be stressed that for alpha spectrometry the number of data was limited, cannot be considered fully representative. In case of gamma-ray spectrometry, large number of data was available but the different data sets were not subjected to detailed statistical analysis (e.g. for outliers) or not sorted the measurement technique further which could help identifying more reproducible gamma-ray spectrometry methods and practices.

8 Conclusions

This report focused on the analysis of two questionnaires linked to the REM2020 proficiency test exercise. The first had to be filled out by participants at the time of registration and the second one at the time of reporting of the measurement results.

In total 108 organisations submitted 117 questionnaire files at the time of registration. Since some participants planned to apply multiple measurement methods they submitted separate questionnaire files for each method. At the reporting stage, JRC-Geel received 89 questionnaire files from 85 laboratories.

In both questionnaires, participants had to fill in 50 fields that contained a single question or some with additional sub-questions. In certain cases, participants could answer in free-text format. Summing up the total number of submitted data, the registration- and reporting surveys included 6341 and 12204 filled-in spreadsheet data cells, respectively.

The participating organisations were mainly laboratories 1) monitoring radioactivity in the environment, 2) research and development and 3) governmental laboratories. Around 25% of the participants were from universities or monitoring of nuclear facilities.

Approximately 60% of the participants measure naturally occurring radionuclides in building materials. The total number of building material samples measured annually was between 1 and 400, with a median of 20 samples. The years of experience varied between 0 to 60 years (average 20 years). Among the participants, 22 are having legal obligations to perform these types of measurements due to national regulations accreditations or contracts.

Regarding samples and sample preparation, about half of the laboratories perform moisture content determination in the building material samples as part of their standard procedure. However, in the REM2020 PT, 81 out of 85 participants (about 95%) who reported measurement results determined the moisture content of all three PT samples.

More than half of the laboratories (56%) receive building material samples as pre-treated samples while others receive samples as produced. The most common processing techniques were crushing, homogenising, sieving, milling or tapping/compacting. When sample is sieved, the particle fractions ranged varies from 63 μm up to 90 mm.

As expected, the most labour intense PT sample was NORM-02 (expanded clay blocks) that was distributed to the participants as one large piece of sample and the material had to be further processed. It was reported that most of the aforementioned sample processing techniques was applied on NORM-02 material to achieve the particle size required for the standard measurement protocol. About 22 participants encountered difficulties while processing or measuring NORM-02 PT reference material.

As indicated in the reporting questionnaire, all 85 participants followed the standard procedure during measurement of PT materials. However, four participants reported minor changes in their procedures when REM2020 PT samples were measured.

The following naturally occurring radionuclides are measured in the laboratories (taken from the registration survey): K-40 (104 participants), Ra-226 (93 participants), Th-232 (74 participants), U-238 (74 participants), Pb-210 (66 participants), Ra-228 (64 participants), Th-228 (59 participants), others (31 participants).

The most frequently used measurement techniques were gamma-ray spectrometry (94 participants), and alpha-particle spectrometry (12 participants), followed by ICP-MS (2 participants). The rest of the techniques (ICP-OES, LSC, solid scintillation counting) were used by one participant each.

The efficiency calibration approaches for gamma-ray spectrometry were mainly based on 1) reference material/source (65 participants), 2) Monte Carlo calculations (15 participants) or 3) combination of Monte Carlo calculations and reference material/source measurements (14 participants).

For alpha-particle spectrometry calibration some participants reported the use of mixed sources, reference materials and also radioactive tracers. The ICP based methods applied multiple-point linear calibration using standard solution.

In 24 cases the containers were not filled completely for gamma-ray spectrometry measurements, of which 21 participants did not fill the void with any filling materials at all.

Regarding detection limits, the median detection limit values were below 10 Bq/kg for all seven radionuclides. However, for some radionuclides very high and wide range of detection limit values were submitted. The best examples are Ra- 226 and K-40 with 15- 1000 Bq/kg and 50-5000 Bq/kg of detection limits, respectively. Reporting such uncertainties for this type of samples and radionuclides is a clear indication of technical issues. It is possible that the method is not under control or possibly there could be a misunderstanding when it comes to the (often complex) detection limit calculation. It can also happen that the decimals/measurement units are not correctly given (so-called “blunders”) or the combination of the listed possible reasons.

When calculating the detection limits, Currie's method (Currie, 1968) and ISO 11929 (ISO, 2019) were applied by 39 and 38 participants, respectively.

The median of the submitted uncertainties in ascending order is the following:
K-40 < Ra-228 < Ra-226 < Th-232 < Th-228 < U-238 < Pb-210.

The spread of uncertainties follows almost the same order, where K-40 has the lowest range while U- 238 and Pb-210 show the highest variation in uncertainties followed by Ra-228.

On the basis of the questionnaires and performance scores, a general best sample preparation practice for gamma-ray spectrometry could be proposed as the following:

- Density and (ideally) chemical composition determination,
- Moisture determination and/or drying at 105 °C until constant weight,
- Processing (e.g. sieving, milling crushing)
- Homogenising,
- Filling a test portion into the measurement container with a well-defined geometry,
- Weighing,
- Adding filling material (e.g. paraffin) if not filled the container fully,
- closing the measurement container radon-tight (radon tightness should be verified),
- Storage before measurement: until radioactive equilibrium between Ra-226 and its decay products is achieved (general practice 14-30 days).

The alpha spectrometry procedures did not differ from each other as much as some steps of the gamma-ray spectrometry procedures. The analytical sequence of alpha-spectrometry started with total digestion of samples with acids or ashing in a muffle furnace in the presence of yield tracers. This step was followed by preconcentration of the radionuclides of interest and chemical separation. Pre-concentration of actinides was done by dissolution of dried residue, $Fe(OH)_3$ co-precipitation or liquid-liquid solvent extraction using tributyl phosphate (TBP). Separation/purification was performed on anionic exchange resin and extraction chromatographic resin and eventually alpha counting sources were prepared by co-precipitation or electrodeposition.

The situation could be considered statistically representative for gamma-ray spectrometry only and with some restrictions for alpha-particle spectrometry. For liquid scintillation counting, solid scintillation counting and ICP techniques as the number of laboratories using these techniques were limited comparing to the participants using gamma-ray spectrometry technique, it was not possible to make a representative data analysis. However, it has to be noted that these techniques are more frequently used in the analysis of less complex samples (e.g. water, air filters) with sufficient accuracy and performance characteristics. Also, more and more laboratories have access to state-of-the-art mass spectrometry based coupled techniques (ICP-MS, ICP-OES etc.) so PT organisers should also focus on their performance in the future.

The second largest data sets, with 77 submitted results, were obtained for alpha-particle spectrometry. From the Youden plots for method repeatability and the reproducibility standard deviation by comparing the between laboratory results, it can be stated that the performance of alpha spectrometry measurement method was very robust, method reproducibility and repeatability were sufficient. For certain radionuclides the method showed better reproducibility than gamma-ray spectrometry. Alpha-particle spectrometry method proved to be accurate and can still claim its place in the radioanalytical toolkit in certain situations as a standalone- or complementary method, despite its shortcomings like resource demands, relatively time consuming- and often tedious procedures.

From the participants' scores and evaluation of the on-line questionnaires a general conclusion was that the performance of laboratories in this PT was satisfactory. The majority of the laboratories managed to submit measurement results within the pre-defined standard deviation range for all seven radionuclides in the three PT materials (see z score). Method accuracy is a problem only for a few gamma-spectrometry laboratories while it may be more of an issue at ICP-OES, LSC laboratories measuring these kinds of samples. However, there is an increased concern related to uncertainty budgets as almost 30% of the zeta scores were either questionable or unsatisfactory. The analysis of questionnaires also confirmed that in certain cases uncertainty budget may not be established and understood properly.

It was an interesting observation that none of the participants submitted measurement results from different measurement methods for the same radionuclide. Either it was because lack of time to perform apply multiple analytical methods or the routine measurement techniques were validated for certain radionuclides.

A simple simulation was performed on how the scores would change when more stringent standard deviations for proficiency assessment (σ_{pt}) were introduced. After decreasing the σ_{pt} from 20% to 15% for K-40, Ra-226, Ra-228, Th-228 and Th-232 and for U-238 and Pb-210 σ_{pt} from 30% to 20%, it was found that the performance scores did not deteriorate dramatically. The total number of acceptable results decreased just by few percentage points for the majority of radionuclides. The overall success rate of z score dropped from 94.7% to 90.2%.

Therefore, the following modified relative standard deviations for proficiency assessment could be used in the next similar proficiency test:

— 15% for K-40, Ra-226, Ra-228, Th-228 and Th-232,

— 20% for U-238 and Pb-210.

References

Currie, L.A. (1968). Limits for qualitative detection and quantitative determination. Application to radiochemistry. Anal. Chem., 40, 586–593.

Hult, M., Jobbágy, V., Sobiech-Matura, K., (2019). Radioactivity monitoring: How the JRC verifies results from monitoring within the European Union, EUR 29789 EN, Publications Office of the European Union, Luxembourg, 2019, ISBN 978-92-76-08762-5, doi:10.2760/124550, JRC117258.

Hult, M., Sobiech-Matura, K., Robouch, P., Emteborg, H., Marissens, G., Stroh, H. and Malo, P., (2024a). REM 2020 proficiency test on naturally occurring radionuclides in building materials, Publications Office of the European Union, Luxembourg, 2024, https://data.europa.eu/doi/10.2760/0952660, JRC135096.

Hult, M. and Paepen, J., (2024b). Outcome of workshop and training on radioactivity in building materials, Publications Office of the European Union, Luxembourg, 2024, JRC140182.

ISO 13528:2015: Statistical methods for use in proficiency testing by interlaboratory comparison, International Organization for Standardization, Geneva.

ISO 5725-2:2019: Accuracy (trueness and precision) of measurement methods and results Part 2: Basic method for the determination of repeatability and reproducibility of a standard measurement method. International Organization for Standardization, Geneva.

ISO 11929:2019 Part 1-3: Determination of the characteristic limits (decision threshold, detection limit and limits of the confidence interval) for measurements of ionizing radiation — Fundamentals and application. International Organization for Standardization, Geneva.

Lozano, J.C., Fernandez, F. and Gomez, J.M.G., (1997). Determination of radium isotopes by BaSO4 coprecipitation for the preparation of alpha-spectrometric sources. J. Radioanal. Nucl. Chem., 223, pp. 133–137.

Paepen, J., Van Ammel, R., Hult, M., Marissens, G., Stroh, H., Emteborg, H., Seghers, J., (2023a). Materials used in the collaborative assessment experiment for the validation of the precision of CEN/TS 17216. JRC-Geel, 2023, JRC135452.

Paepen, J., Van Ammel, R., Hult, M., Marissens, G., Stroh, H., Emteborg, H., Seghers, J., (2023b). NORMCONSTRUCT: Results of the collaborative assessment experiment for the validation of the precision of CEN/TS 17216. JRC-Geel, 2023, JRC135812.

Sill, C.W., (1987). Determination of Ra-226 in ores, nuclear wastes and environmental samples by high resolution α-spectrometry, Nucl. Chem. Waste Manage., 7, pp. 239-256.

Youden WJ (1959). Graphical diagnosis of interlaboratory test results. Industrial Quality Control, 15, 24-28.

List of abbreviations, acronyms and initialisations

Abbreviations	Definitions
BGO crystal	Bismuth Germanium Oxide (scintillation) crystal
CEN	European Committee for Standardization
CRM	Certified Reference Material
DG-ENER	Directorate General for Energy (of the European Commission)
HPGe detector	High-Purity Germanium detector
IAEA	International Atomic Energy Agency
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
ICP-OES	Inductively Coupled Plasma - Optical Emission Spectroscopy
IEC	International Electrotechnical Commission
inox	Stainless steel, known as inox in French and some other languages
ISO	International Organization for Standardization
JRC	Joint Research Centre
KRISS	Korea Research Institute of Standards and Science
LSC	Liquid Scintillation Counting
LVS	Latvian standard
MC-ICP-MS	Multicollector - Inductively Coupled Plasma - Mass Spectrometer
NaI	Sodium-iodide. A material used as scintillation detector
NORM	Naturally Occurring Radioactive Material
PT	Proficiency Test

Abbreviations	Definitions
REM	Radioactivity Environmental Monitoring
STN	Slovak standard
TBP	TriButyl Phosphate
TENORM	Technologically Enhanced NORM
TCS	True Coincidence Summing
UNI	Italian standards body

List of figures

List of tables

Annexes

Annex 1. Questionnaire-1: at the time of registration

REM 2020 PT Survey on the measurement methods

Fields marked with * are mandatory.

Dear participant of the REM 2020 PT on naturally occurring radionuclides in building materials,

We kindly ask you to answer questions regarding the procedures used in your laboratory to measure naturally occurring in building materials.
Questions marked with asterisk (*) are mandatory.

Contact details

* Contact person
---

* Full email of the contact person
test@test.com

* Name of the institution
---

* What is the type of your laboratory? (More than one answer possible)
- [ ] Research and development
- [x] Monitoring radioactivity in the environment
- [ ] Monitoring nuclear facilities
- [ ] Governmental laboratory
- [ ] University laboratory
- [x] Other

* Please specify

Samples of building materials containing naturally occurring radionuclides

1

* Do you measure naturally occurring radionuclides in building materials in your laboratory?
- (•) Yes
- ( ) No

* What kind of material do you usually measure?
---

* Approximately, how many building material samples per year do you measure?
0

* How many years of experience does your laboratory have in performing this kind of measurements?
0 years

* Do you perform moisture content determination in these samples?
- ( ) Yes
- (•) No

* In which form do you receive the building materials? (More than one answer possible)
- [x] As produced (e.g. whole bricks, aggregates)
- [ ] Pre-treated (e.g. pulverised concrete)

* What kind of processing do you apply during sample preparation? (Please choose all relevant answers, more than one aswer possible)
- [x] Crushing
- [x] Milling
- [x] Sieving
- [x] Homogenising
- [x] Tapping/Compacting
- [x] Other
- [ ] None

* What kind of tools/apparatus do you use for crushing?
---

* What kind of tools/apparatus do you use for milling?

2

* What kind of tools/apparatus do you use for sieving?
---

* Please provide the mesh sizes of the sieves used
---

* Please provide the typical most representative fraction, average and end point used for sieving
---

* What kind of tools/apparatus do you use for homogenising?
---

* What kind of tools/apparatus do you use for tapping/compacting?
---

* Please provide more information
---

* Are you legally obliged to perform these measurements?
- (•) Yes
- ( ) No

* By whom?
---

* Please provide the legal basis for this obligation

3

Measurements methods

* Which of the following naturally occurring radionuclides are routinely measured in you laboratory (in any kind of material)? (More than one answer possible)
- [x] K-40
- [x] Pb-210
- [x] Ra-226
- [x] Ra-228
- [x] Th-228
- [x] Th-232
- [x] U-238
- [x] Other

* Please specify the radionuclide(s)
---

* Which method is used for the measurement of K-40?
- (•) Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- ( ) Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ( ) By determination of total potassium
- ( ) LSC
- ( ) Other

* Which method is used for the measurement of Pb-210?
- ( ) Alpha-particle spectrometry
- ( ) Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- ( ) Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ( ) ICP-MS
- ( ) LSC
- (•) Other

* Please specify the method
---

* Which method is used for the measurement of Ra-226?
- ( ) Alpha-particle spectrometry
- ( ) Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- ( ) Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ( ) ICP-MS
- ( ) LSC

4

- (●) Other

* Please specify the method

---

* Which method is used for the measurement of Ra-228?
- ( ) Alpha-particle spectrometry
- ( ) Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- ( ) Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ( ) ICP-MS
- ( ) LSC
- (●) Other

* Please specify the method

---

* Which method is used for the measurement of Th-228?
- ( ) Alpha-particle spectrometry
- ( ) Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- ( ) Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ( ) ICP-MS
- ( ) LSC
- (●) Other

* Please specify the method

---

* Which method is used for the measurement of Th-232?
- ( ) Alpha-particle spectrometry
- ( ) Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- ( ) Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ( ) ICP-MS
- ( ) LSC
- (●) Other

* Please specify the method

---

* Which method is used for the measurement of U-238?
- ( ) Alpha-particle spectrometry
- ( ) Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)

5

- ( ) Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ( ) ICP-MS
- ( ) LSC
- (●) Other

* Please specify the method

---

* You indicated additional radionuclide(s), not listed in the first question. Which method is used for the measurement of this radionuclide(s)?
- ( ) Alpha-particle spectrometry
- ( ) Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- ( ) Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ( ) ICP-MS
- ( ) LSC
- (●) Other

* Please specify the method

---

* Please describe briefly the sample preparation, if any

---

* Which type of efficiency calibration do you use for gamma-ray spectrometry?
- ( ) Reference material or source
- ( ) Monte Carlo calculations only
- ( ) Combination of Monte Carlo calculations and reference material or source measurements
- (●) Other

* Please provide more details on the efficiency calibration used

---

* Is the measurement container for gamma-ray spectrometry completely filled with the sample?
- ( ) Yes
- (●) No

* Are there any fillers applied to fill the void?
- (●) Yes
- ( ) No

6

* Please provide additional information on the type of fillers used

---

* Do you apply any sealing material to close the measurement container for gamma-ray spectrometry?
- (●) Yes
- ( ) No

* What kind of sealing it do you apply to close the measurement container?

---

* Do you test the radon tightness of the measurement containers used for gamma-ray spectrometry?
- (●) Yes
- ( ) No

* Please provide a brief description of this method

---

* Please describe the calibration procedure(s) used for your measurement method(s) (other than gamma-ray spectrometry)

Problems encountered

* Do you encounter any difficulties while measuring the naturally occurring radionuclides in building materials?
- (●) Yes

7

- ( ) No
- ( ) Not applicable

* Please describe these difficulties

Contact

Katarzyna.SOBIECH-MATURA@ec.europa.eu

8

Annex 2. Questionnaire-2: at the time of reporting

REM 2020 PT Survey on the measurement methods

Fields marked with * are mandatory.

Dear participant of the REM 2020 PT on naturally occurring radionuclides in building materials,

We kindly ask you to answer questions regarding the procedures used during measurements of the PT reference materials.
Questions marked with asterisk (*) are mandatory.

Contact details

* Contact person

---

* Full email of the contact person

test@test.com

* Name of the institution

General information on the performed measurements

* Did you follow your standard procedure during the measurement of the PT reference materials?

- Yes
- (•) No

* What type of changes did you make?

---

* Did you determine the moisture content of the the samples according to the provided procedure?

1

- Yes
- (•) No

* What type of changes did you make?

What was the moisture content of the reference materials? (in mass %)

	Moisture content (%)
NORM1	0
NORM2	0
NORM3	0

* What kind of processing did you apply during sample preparation of NORM1? (Please choose all relevant answers, more than one answer possible.)

- [ ] Crushing
- [ ] Milling
- [ ] Sieving
- [ ] Homogenising
- [ ] Tapping/Compacting
- [x] Other
- [ ] None

* Please provide more information

---

* What kind of processing did you apply during sample preparation of NORM2? (Please choose all relevant answers, more than one answer possible)

- [x] Crushing
- [x] Milling
- [x] Sieving
- [x] Homogenising
- [x] Tapping/Compacting
- [x] Other
- [ ] None

* What kind of tools/apparatus did you use for crushing?

2

---

* What kind of tools/apparatus did you use for milling?

---

* What kind of tools/apparatus did you use for sieving?

---

* Please provide the mesh sizes of the sieves used

---

* What kind of tools/apparatus did you use for homogenising?

---

* What kind of tools/apparatus did you use for tapping/compacting?

---

* If other, please provide more information.

---

* What kind of processing did you apply during sample preparation of NORM3? (Please choose all relevant answers, more than one answer possible.)

- [ ] Crushing
- [ ] Milling
- [ ] Sieving
- [ ] Homogenising

3

- [ ] Tapping/Compacting
- [x] Other
- [ ] None

* Please provide more information

Measurements methods

* Which of the following naturally occurring radionuclides did you measure in the REM 2020 PT reference materials? (More than one answer possible)

- [x] K-40
- [x] Pb-210
- [x] Ra-226
- [x] Ra-228
- [x] Th-228
- [x] Th-232
- [x] U-238
- [x] Other

* Please specify the radionuclide(s)

---

* Which method was used for the measurement of K-40?

- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- By determination of total potassium
- LSC
- (•) Other

* Please specify the method

---

* Which method was used for the measurement of Pb-210?

- Alpha-particle spectrometry
- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ICP-MS
- LSC
- (•) Other

4

* Please specify the method

---

* Which method was used for the measurement of Ra-226?

- Alpha-particle spectrometry
- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ICP-MS
- LSC
- Other

* Please specify the method

---

* Which method was used for the measurement of Ra-228?

- Alpha-particle spectrometry
- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ICP-MS
- LSC
- Other

* Please specify the method

---

* Which method was used for the measurement of Th-228?

- Alpha-particle spectrometry
- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ICP-MS
- LSC
- Other

* Please specify the method

---

* Which method was used for the measurement of Th-232?

- Alpha-particle spectrometry
- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ICP-MS

5

- LSC
- Other

* Please specify the method

---

* Which method was used for the measurement of U-238?

- Alpha-particle spectrometry
- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ICP-MS
- LSC
- Other

* You indicated additional radionuclide(s), not listed in the first question. Which method was used for the measurement of this radionuclide(s)?

- Alpha-particle spectrometry
- Gamma-ray spectrometry with high resolution detectors (e.g. HPGe)
- Gamma-ray spectrometry with low resolution detector (e.g. NaI)
- ICP-MS
- LSC
- Other

* Please specify the method

---

* Please describe briefly the sample preparation, if any

---

* What was the volume of the measurement container for the gamma-ray spectrometric measurements? (in litres)

0 L

* What was the mass of the sample used for the gamma-spectrometric measurements? (in grams)

0 g

* Which type of efficiency calibration did you use for gamma-ray spectrometry?

- Reference material or source
- Monte Carlo calculations only
- Combination of Monte Carlo calculations and reference material or source measurements

6

- Other

* Please provide more details on the efficiency calibration used

---

* Were the measurement containers for gamma-ray spectrometry completely filled with the sample?

- Yes
- No

* Were there any fillers applied to fill the void?

- Yes
- No

* Please provide additional information on the type of fillers used

---

* Did you apply any sealing material to close the measurement container for gamma-ray spectrometry?

- Yes
- No

* What kind of sealing did you apply to close the measurement container?

---

* Please describe the calibration procedure(s) used for your measurement method(s) (other than gamma-ray spectrometry)

7

Please provide values for detection limits of the radionuclides measured. If one or more radionuclides was not measured in your laboratory please insert N/A.

	NORM1	NORM2	NORM3
K-40	0	0	0
Pb-210	0	0	0
Ra-226	0	0	0
Ra-228	0	0	0
Th-228	0	0	0
Th-232	0	0	0
U-238	0	0	0
Other	0	0	0

8

Annex 3. Brief description of the sample preparation as reported in the registration survey

- Acid digestion in closed vessel (4 acids, assisted microwave) + separation by ion exchange resin
- Spiking with U-236 Th-229, dissolve in a mixture of acids, separate using anion resins in columns
- Closed vessel acid digestion in a mixture of HNO3 - HF and H2O2, evaporation of the solution and take up in 0.45M nitric. Measure U, Th and K by ICP-OES and measure a dilute fraction by ICP-HRMS.
- the sample is added to the Marinelli vessel and is measured after 30 days
- weighing into a Marinelli container
- shaking and homogenization
- 1/ crushed and milled if necessary, 2/ put in a container, 3/ gas tightness bag, 4/ +/-30 days waiting time, 5/ measurement.
- weight, dry (if needed), prepare for our geometry
- put the sample in our calibrated geometry
- as it is or drying
- A 2% addition of charcoal powder is added regarding to trap Rn into the sample volume.
- crushing milling homogenization oven-dried sealing
- Crushing, homogenisation, weighting
- Grind, sieve, calcine and digest
- Acid digestion with HF and HClO4 + HNO3 on PTFE beaker
- Acid digestion with HF and HClO4 and ICP-MS measurement
- The accredited method is for testing the material in its intended form of use
- Firstly, the sample is dried and its moisture is determined. Then, the sample is milled and homogenized. Finally, the sample is compacted until reaching the desired thickness.
- If it is required, we grind the sample and condition it for 14 days near the measuring device.
- For water samples we just measure 500 mL in a volumetric cylinder and put it in a pre-weighed Marinelli beaker. For foods we use a blender to homogenize the food and put it in a Marinelli beaker so that the density is 1.
- dry, crush, sieve, homogenize, fill geometry
- placing the materials in the Marinelli cup
- We use sealed containers
- Drying, grinding, homogenizing and sieving.
- It depends on the sample type

- the sample is homogenized and its apparent density is verified to be between 0.7 kg/L and 1.3 kg/L. It is then placed in a 1 L Marinelli beaker
- Drying, grinding, homogenizing and sieving.
- crashing; sieving; drying; homogenizing
- The samples are homogenized and packed in the standard boxes of 260 ml or 100 ml.
- Drying, grinding, homogenizing and sieving.
- No preparation. if necessary (if the sample has been heated or subjected to mechanical actions), we wait for the Ra 226 to be in equilibrium with the progeny (almost 20 days). In building materials samples, the loss of Radon 222 and subsequent loss of equilibrium of the Ra226 chain is almost negligible
- none
- Sample is dried, crushed, grinded to <2 mm and (sometimes) milled <1 mm
- ICP-MS and Alpha-particle spectrometry: Acid digestion and separation using ion exchange resins TEVA, TRU, UTEVA, etc, Gamma-ray spectrometry: Drying and spraying of samples
- no preparation
- samples were dried, milled, homogenised, filled in beakers, gas-tight, set for ~23 days to allow equilibrium in Ra-226 decay chain
- sample is shredded and sealed in a stainless-steel beaker
- Pretreatment: Sample drying-Grinding(pulverizing)-homogenization-bottling, separation and purification: Radiochemistry, source preparation: electrodeposition or micro-coprecipitation
- For water: slow evaporation with infrared bulbs, dry residue, ground, sieved, encapsulated in geometric boxes known for spectrum acquisition. For soil: bringing to constant mass, encapsulation in a box with known geometry, maintaining for 3 weeks for secular balance, until a first spectrum acquisition.
- homogenization, filling into a measuring vessel, weighing
- for construction materials no prior preparation is performed
- Sample is measured in 0.5 or 1.0 L Marinelli beaker
- Sieved and homogenized pulverised material, placed in a sealed beaker.
- sieving and drying
- drying, vacuuming for 3 weeks
- drying, crushing, sieving, packing to Marinelli beaker and determination of net mas of sample
- The sample is dried in a laboratory oven at 105 °C. It is crushed and sieved (2 mm), hermetic closed in a Marinelli container (volume 450 mL) or measurement container (80 mL). The sample is measured 30 days after hermetic closed.

- The liquid samples are evaporated to dryness, then the dry residue is ground and placed in the measuring geometry. The solid samples are dried at a constant mass, and then they are ground and homogenized and placed in the measuring geometry (cylindrical plastic box).
- Grinding preparation and humidity measurement
- Drying, homogenization and weighing
- Direct measurement (dry powder samples) or pre-drying and crushing if necessary.
- The samples are dried and brought to a constant mass; then the material is sieved until it reaches the defined grain size (<2 mm).
- Using Marinelli containers
- we receive the samples already ground
- ashing at 600 °C and acid digestion
- filling to cylindrical container, sealing container, equilibration, measurement
- Alpha emitting radionuclides are extracted from matrices using leaching and chromatographic separation
- As described in the UNI 11665:2017
- Using cylindrical container which is same geometry with calibration standard.
- Our samples are transferred to 450 mL standard Marinelli beaker geometry
- Samples have to be grinded (if they are not yet) and homogenized before transferring in a calibrated container.
- the samples is crushed, sieved, homogenized, cylindrical geometry is used
- If needed material is crushed by hammer and milled in the ceramic mill or mortar and pestle, if needed the material is dried at 105 °C. The material is sieved and then placed into cylindrical beaker up to the top, then closed and sealed using elastic duct tape.
- Drying, milling
- Homogenize, dry, sieve, and fill the beaker, compact and seal with laboratory film.
- solid sample is dried, crushed, milled, sieved and homogeneized. 107.5 mL of this material is introduced into a plastic cylindric container. This geometry is simulated by using Labsocs from Canberra industry.
- N.a.
- We try to keep sample preparation to a minimum for gamma spectrometry. But if necessary, we grind, mix and take subsamples.
- drying, crushing, sieving, packing to Marinelli beaker and determination of net mas of sample
- The samples are dried, crushed and sifted.
- Drying until constant weight is reached.
- drying, mill, sieving, put into the container, sealing

- Drying, sieving, crushing, milling, homogenising and reducing, packing in a measuring vessel.
- For alpha spectrometry After acid digestion, or leaching process, Ion exchange resin AGx8 and UTEVA resin are used. Finally, an electrodepositon method is carried out for the source preparation
- The sample is dried (if necessary) in an oven and prepared in a sealed container
- If it is not pre-prepared, the sample is crushed and homogenized before measurement in a calibrated geometry
- The sample is dried (if required) in an oven and is calcined (if required) and prepared in a sealed container.
- The soil samples are dry, homogenized and put in the cylindrical boxes
- About fruit and vegetables they are blended with a mixer to homogenize the sample. So the product is put in a Marinelli Beaker, ready to measure. About the soil, the operations are: drying, grinding and sieving.
- none
- no
- Essiccazione, setacciatura a 2 mm e omogeinizzazione (translation to English by author: “Drying, sieving at 2 mm and homogenisation”)
- The material is dried at 105 degrees C overnight and the sample is put into a metal container. The container is sealed with plasticine and isolation tape.
- U-238; 0.25ml conc. HNO_3 added to 50ml water. Gamma spectrometry; Samples are oven dried, sieved through a 2mm sieve and homogenised. An aliquot is placed in a well-defined counting geometry and then measured on a high-resolution gamma spectrometer. Appropriate corrections (TCS and self-attenuation) are applied. Results are quoted on a dry weight basis.
- Minimal sample preparation is usually required (homogenization, compliance with calibration geometry, …)
- The samples are dried at 100 °C overnight, and packed in cylindrical container (100 or 200 mL) or in Marinelli beakers (1000 mL). The containers are sealed with tape and left for at least 30 days for the equilibrium.
- If the sample is solid, we crush or ground it, then homogenize it to the fine powder and put into containers (Marinelli 05 l or Marinelli 1 L), for measurement.
- Grinding preparation and humidity measurement
- Internal method
- As describes previously
- the sample is crushed, dried and homogenized. The samples were weighed and transferred to a Marinelli beaker (500 mL capacity), carefully sealed, and stored for four weeks to allow for secular equilibrium between thorium, radium and their decay products.
- None

- described beforehand
- an aliquot of the sample is placed in a measuring geometry - the sample container is filled up to the top and then sealed inside an aluminium foil
- After sieving to 2 mm. sample packaged in a plastic bottle of standard geometry (100 ml, 250 ml or 500 ml). Addition of 10% by volume of activated carbon during the preparation for the measurement of Ra-226 carried out after 30 days of equilibration.
- alpha spec: extraction chromatography + electrodeposition, LSC: liquid/liquid extraction by selective cocktail
- An aliquot of the sample is placed in the sample container. The sample container is filled up to the top.
- The sample is dried at 105 deg.C until the mass is constant.
- Radiochemical separation for alpha spectrometry
- Fill the measuring container with the pulverized sample Close the container and wait 21 days for the gamma measurement
- Drying, grinding, packaging and sealing for secular equilibrium in case of gamma spectrometry measurements. Radiochemistry in case of alpha spectrometry (U, Th and Po isotopes).
- for alpha particle spectrometry: calcination and column separation is done
- In the case of soils samples are dried. Ra-228 is determined using Ac-228 of 911.2 keV; Ra-226 is determined using Pb-214 of 351.9 keV and U-238 is determined using Pa-234m of 1001.03 keV (if any).
- for Ra-226 air-tight sample shielding to attain equilibrium with short-lived radon daughters
- After crushing and drying the sample , we put it in a cylinder or Marinelli beaker radonproof
- dry, crush, sieve, put in measurement beaker
- Sample drying at 40°C, sieving through 2mm and drying at 110°C. For gamma spectrometry, we add another step: sieving through 0.5mm.
- Omogeneizzazione del campione e travaso nel corretto contenitore di Marinelli, in alcuni casi essicazione del campione o determinazione del secco (Translation by author: “homogenization of the sample and decanting into the correct Marinelli container, in some cases drying of the sample or determination of the dryness”)
- "The sample is homogenized with mortals, ball-mill or mixer depending on the properties of the sample. If Ra-226 is measured the geometry is fully filled and sealed with radon tight plastic in a vacuum chamber. We only have calibration for density 1.0 g/cm3 for fully filled geometries.
- If the density of the sample is closer to 1.5 the other nuclides are measured with a geometry that isn't fully filled using a calibration source of 1.5 g/cm3."
- - hermetization (at least for 23 days) - STN EN ISO 18589-3 (JRC’s/Authors’ comment: “hermetically sealed”)

Getting in touch with the EU

In person

All over the European Union there are hundreds of Europe Direct centres. You can find the address of the centre nearest you online (european-union.europa.eu/contact-eu/meet-us_en).

On the phone or in writing

Europe Direct is a service that answers your questions about the European Union. You can contact this service:

— by freephone: 00 800 6 7 8 9 10 11 (certain operators may charge for these calls),

— at the following standard number: +32 22999696,

— via the following form: european-union.europa.eu/contact-eu/write-us_en.

Finding information about the EU

Online

Information about the European Union in all the official languages of the EU is available on the Europa website (european-union.europa.eu).

EU publications

You can view or order EU publications at op.europa.eu/en/publications. Multiple copies of free publications can be obtained by contacting Europe Direct or your local documentation centre (european-union.europa.eu/contact-eu/meet-us_en).

EU law and related documents

For access to legal information from the EU, including all EU law since 1951 in all the official language versions, go to EUR-Lex (eur-lex.europa.eu).

EU open data

The portal data.europa.eu provides access to open datasets from the EU institutions, bodies and agencies. These can be downloaded and reused for free, for both commercial and non-commercial purposes. The portal also provides access to a wealth of datasets from European countries.

Science for policy

The Joint Research Centre (JRC) provides independent, evidence-based knowledge and science, supporting EU policies to positively impact society

EU Science Hub
Joint-research-centre.ec.europa.eu

Publications Office
of the European Union