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Radioactivity monitoring: How the JRC verifies results from monitoring within the European Union

— A quick guide

A brief introduction to proficiency tests and how they are used to make sure EU citizens are better protected from harmful levels of radioactivity. In support of Euratom Treaty Articles 35 and 36.



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Contact information

Name: Mikael Hult

Email: mikael.hult@ec.europa.eu

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Contents

A brief background.....	2
About this quick guide.....	7
Brief description of 10 proficiency tests between 2003 and 2018.....	9
Overview.....	10
PT I, Air filter (2003).....	12
PT II, Milk powder (2005).....	14
PT III, Mineral water (2008).....	16
PT IV, Soil (2010).....	18
PT V, Dried bilberries (2011).....	20
PT VI, Mineral water (2012).....	22
PT VII, Air filter (2014).....	24
PT VIII, Air filter (2016).....	26
PT IX, Dried maize (2017).....	28
PT X, Drinking water (2018).....	30
Future directions.....	32
Annex: Introduction to radioactivity measurement and the activities at JRC-GEEL — A brief tutorial.....	35
Measuring radioactivity.....	36
Why we measure radioactivity.....	38
Case studies.....	40
Activities at JRC-Geel.....	42
JRC-Geel's core competence: proficiency tests.....	44
List of initialisms.....	47

A brief background

Why is the European Commission overseeing the radioactivity monitoring of the EU Member States?

According to Chapter 3 of the Euratom Treaty (i.e. Articles 30-39), the EU Member States are responsible for the implementation of basic standards laid down within the Community for the protection of the health of workers and the general public against the dangers arising from ionising radiations. Articles 35 and 36 oblige the Member States to monitor the level of radioactivity in the air, water and soil and to report to the European Commission. The Euratom Treaty also obliges the Commission to verify the operation and efficiency of this monitoring. The purposes of the monitoring are the following.

- Ensuring the health and safety of citizens (ensuring basic safety standards).
- Providing accurate monitoring results to the national radioprotection agencies and the Commission so they have the necessary data to enable them to trigger or recommend additional health and safety measures due to an incidental or intentional release of radioactivity in the environment, for example a nuclear accident or a malicious act.

Today, much of the data is also made publicly available by the Commission's Joint Research Centre (JRC). This brings added value as the international scientific community can find information for itself, which can, for example, be used to prove the validity of environmental dispersion models.

What are the tasks of the JRC?

The JRC supports the Commission's Directorate-General for Energy with technical and scientific input into its work. The JRC does so by verifying the Member States' implementation of Chapter 3 of the Euratom Treaty in numerous ways. **Note that this report deals (solely) with the specific task of assessing the quality of the radiological monitoring results that the Member States report to the Commission.**

Who is doing this job?

A multinational expert team at the JRC, most of them with a master's degree or a PhD.

Where and when does it take place?

At JRC-Geel in northern Belgium, where the quality of the data from the Member States' monitoring programmes has been continuously assessed since 2003.

What are proficiency tests and how are they used?

To verify the procedures and methods of the monitoring labs of the Member States, 10 proficiency tests (PTs) were conducted by the JRC. These took place between 2003 and 2018, and were based on sample materials such as air filters, water, crops and soil. First the JRC produces a reference material that contains a well-established quantity of radioactivity. It then distributes samples to the Member States' labs. Finally the JRC collects the data, compares the results and gives feedback and recommendations.

This document describes all 10 PTs — why and how they were designed, and how they have been used to ensure the quality of the monitoring of environmental radioactivity in the Member States of the EU. The text also provides proposals for improvements in best practices and international standards.

The PTs also serve as a means for certain institutes to show their traceability to the International System of Units (SI). This was possible because the JRC was a signatory of the mutual recognition agreement on specific calibration and measurement capabilities, registered in the database of the International Bureau of Weights and Measures (BIPM) in Paris.

Please note: In this report some aspects and results have been presented in an abbreviated and simplified manner. For detailed scientific-technical reports see <https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Observations and conclusions

GENERAL

- **The general status of radioactivity monitoring in the EU is good.**

The Chernobyl disaster of 1986 served as an alarm bell, demonstrating that radioactivity monitoring in Europe could be vastly improved, as could international cooperation in these matters. Ever since, ambitions, methodologies, instrumentation and international cooperation have been continually upgraded.

- **New opportunities for research arise as monitoring networks grow and instrumentation improves.**

As lower levels of radioactivity can be detected across a greater network of laboratories, the tracing of radioactivity back to (undisclosed) sources is enabled — even if the release is very small. This not only makes it possible to better assess the impact of any radioactive release, it also provides the scientific community with valuable data that is useful, for example, in studying wind patterns, sea currents, ecological processes and the impact of industries.

- **Better sharing and exchanging of best practices.**

Differences in practices exist between laboratories and countries. This can create comparability problems and can be a source of dispute. However, in certain cases it is also an asset that different approaches are available to reach the same result. Moreover, the multitude of radionuclides and the great variety of matrices being monitored offer very different technical challenges, which are difficult for a single laboratory to master. Therefore, the better sharing and exchanging of experience and best practices remains an overall objective.

PROFICIENCY TESTS

- **PTs have become important tools to study the capacity of the European monitoring labs for generating good-quality data.**

This is not only important for the EU as a whole, but also for the Member States (especially non-nuclear Member States). They are now served with an independent evaluation of the status of their national monitoring and receive top-quality reference materials to help validate their own methods.

- **PTs serve as a complement to verification visits.**

A PT will assess every Member State almost every year. This is a convenient and effective complement to the verification visits that the Commission carries out. During such visits only about four Member States (and one or two installations) per year can be assessed.

The reference materials that are provided in connection to the PTs at hand have been shown to be very useful to Member States' laboratories. This is particularly true in the case of short-lived radionuclides for which no certified reference materials exist (e.g. iodine-131, with a half-life of 8 days; radon-222, with a half-life of 3.8 days).

TRAINING AND EDUCATION

- **Skills, competences and workforce availability.**

The importance of retaining critical competences in nuclear science was already being acknowledged by the nuclear community in the 1990s. In recent years a number of studies have been undertaken to examine the concern that nuclear education and training are in decline. The capacity of a laboratory to provide accurate measurements is closely related to the skill and experience of the staff. We have also made this observation on the basis of follow-up workshops, training sessions and responses received to questionnaires. Accordingly, teaching, education and vocational training are now more of a priority than ever.

MONITORING TECHNIQUES

- **Gross counting techniques give very poor results in the PTs.**

Such counting techniques have proved to be unreliable for environmental monitoring. They should be used with caution and only in special cases where relatively good control of the different radionuclides present in the sample is available.

- **The collection and handling of samples introduces special challenges.**

Sampling — including collection, transport, storage and sample preparation — is an art. Increased efforts and research are necessary to better understand and quantify the errors introduced through environmental sampling. This is particularly true for volatile elements (such as iodine) or inert gases (such as radon).

- **There must be routines to avoid serious errors.**

One mistake that is rare but that has dire consequences is to report an incorrect value that deviates from the correct one by a factor of 1 000 or even 1 000 000. This means, for example, writing 'kg' instead of 'g' or 'Bq' instead of 'mBq', or using a decimal comma instead of a decimal point. Such errors are easy to make, and there must be a process in place by means of which every reported value is double- or triple-checked before reporting.

- **International collaboration is an asset.**

Radiation does not stop at borders. Any severe accident or contamination incident that affects a number of countries inside or outside of the EU will require contacts and the sharing of knowledge. It is therefore positive to see that networking and partnering activities between Member States and their neighbours has improved. There are examples of networking activities at international level. For example, the International Atomic Energy Agency (IAEA) is organising a network called Almera (Analytical Laboratories for the Measurement of Environmental Radioactivity). It has a goal similar to the work described in this report, namely to ensure the quality of monitoring data. The Cellar network (Collaboration of European Low-level Underground Laboratories) initiated by the JRC and the Physikalisch-Technische Bundesanstalt (German national metrology institute) promotes technological developments that serve to increase the robustness of monitoring data.

About this quick guide

This guide gives a general introduction to how and why radioactivity monitoring is performed within the European Union. In non-jargon language, it explains what PTs are and how they are designed and managed by the JRC in Geel, Belgium.

In addition, the tutorial in the Annex gives some context to promote understanding and curiosity, for example on radiation biology, nuclear accidents and measurement techniques.

Note that there is also a list of initialisms at the end of the Annex.

The guide represents an abbreviated and simplified compilation of the official PT reports. (See the REMON website: <https://remon.jrc.ec.europa.eu>).

Brief description of 10 proficiency tests between 2003 and 2018

The full reports on these PTs can be downloaded free of charge from:
<https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Overview

Ten proficiency tests have been organised by JRC-Geel since 2003

In this section the 10 PTs organised up to 2018 are presented, with information on why they were performed, which radionuclides were measured and what the outcome was.

In principle, every Member State was represented in each PT, but not always with the same laboratories. One important aspect of the added value of a PT to the Member States' labs is that they receive a high-quality reference material and can use the PT as an important quality-control tool and for obtaining accreditation. This is particularly important for non-nuclear (or smaller) Member States that are generally less well equipped compared to the two major nuclear Member States (France and the United Kingdom), which also organise their own PT schemes.

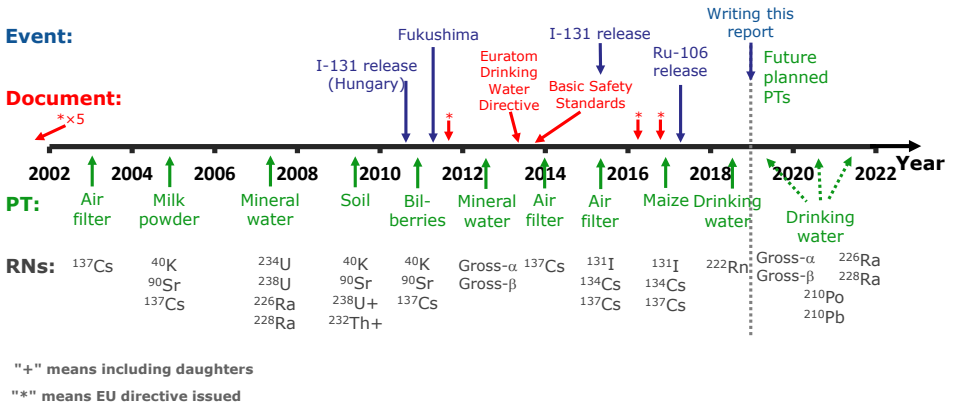
Table 1. Summary of the 10 PTs that have been conducted by the JRC since 2003.

Year	Sample type	Radionuclides	Reporting labs	Reported results (*)
2003	Air filter	^{137}Cs	43	48
2005	Milk powder	^{40}K , ^{90}Sr , ^{137}Cs	63	149
2008	Mineral water	^{226}Ra , ^{228}Ra , ^{234}U , ^{238}U	45	152
2010	Soil	Fifteen different radionuclides	73	743
2011	Dried bilberries	^{40}K , ^{90}Sr , ^{137}Cs	88	222
2012	Mineral water	Gross alpha, gross beta	71	404
2014	Air filter	^{137}Cs	78	76
2016	Air filter	^{134}Cs , ^{137}Cs , ^{131}I	67	201
2017	Dried maize	^{134}Cs , ^{137}Cs , ^{131}I (^{40}K)	120	120
2018	Drinking water	^{222}Rn	101	135

(*) Generally the activity of one radionuclide counts as one result. Not all labs are equipped to measure all radionuclides. In some cases it was agreed that labs could report separate results for a radionuclide using different techniques.

It is the Article 35 and 36 experts from the Member States who suggest to the Commission at their regular meetings which radionuclides and matrices to select for upcoming PTs. This is triggered by issues such as new legislation, developments in instrumentation and the publication of new international standards.

Figure 1. A timeline showing the context of the PTs. It is often EU legislation that drives the need to select certain radionuclides and matrices in the PTs.



Proficiency test I

Air filter (2003)

Highlighting a diversity of lab practices

Why was it performed?

Caesium-137 is a key radionuclide when monitoring radioactive contamination after a nuclear accident, in both the short-term (days) and the long-term (many years, even decades) perspective. Air monitoring is a vital monitoring task, as it has the potential to detect contamination before it enters urban and ecological systems in the immediate aftermath of an accident.

Radionuclides and measurement techniques

Caesium-137 (beta-decaying but also emitting gamma rays; half-life 30 years).

Measurement was based on the gamma rays emitted at 662 keV, detected with a high-purity germanium (HPGe) detector.

Key findings

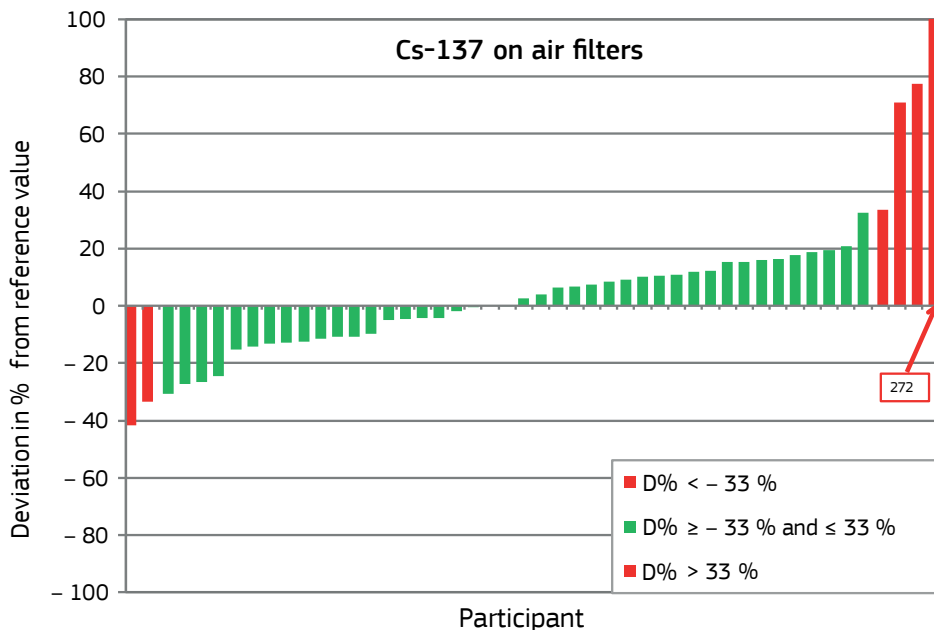
With only a few exceptions, the communication, logistics and cooperation between the JRC and the 43 participating Member State labs worked as planned.

Out of 48 reported measurement results, 42 were within ± 33 % of the JRC-Geel reference value.

Taking both reported value and uncertainty into consideration, 12 labs got unacceptable results; they needed to investigate possible sources of error in their measurement process.

It was revealed that air-sampling procedures differ widely among the Member States, and that the area of the filters used varies by a factor of 2 000. Accordingly, the caesium-137 activity per measured filter varied from 0.03 Bq to 0.6 Bq. Among the 43 participating laboratories, 37 different types of filters were used.

Figure 2. A modest value of $\pm 33\%$ was set in this PT, although caesium-137 in air filters should be relatively easy to measure. The reason for accepting up to 33% relative deviation was that 37 different types of air filters were used by the participating laboratories.



Notes

Caesium-137 is expected to be discharged in large quantities after any major nuclear accident. Accordingly, airborne particles carrying this nuclide serve as an early-warning sign that something out of the ordinary has taken place.

HPGe detectors are the workhorses of routine monitoring laboratories in EU Member States; they are used for most gamma-ray-emitting radionuclides.

In general, filter samples are easy to measure since there is little attenuation of gamma radiation by the filter material. In addition, radiochemical pretreatment is seldom needed.

The full report on this PT can be downloaded free of charge from:

<https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Proficiency test II

Milk powder (2005)

A food source with some analytical challenges

Why was it performed?

It is important to monitor cow's milk for two reasons: it is an important component of human nutrition; and it is sensitive to airborne contamination as a large proportion of the forage for the cows grows in the open air.

A commercially available milk powder reference material with elevated levels of radioactivity was used (IAEA-152). The milk was from cows in Ukraine that have metabolised contaminated fodder from the Chernobyl area.

Radionuclides, radiation and measurement techniques

Caesium-137 (see PT I). With chemistry similar to potassium it has a short biological half-life of about 70 days.

Potassium-40 (naturally present beta emitter; half-life >1 billion years).

Strontium-90 (beta emitter; half-life 29 years). A 'bone-seeking' radionuclide with a complex metabolism in the human body and a very uncertain biological half-life between 18 years and 50 years.

Caesium-137 and potassium-40 were measured using gamma-ray spectrometry (GS), whereas strontium-90 was measured using gas flow proportional counting, liquid scintillation counting (LSC), Cherenkov counting, plastic scintillators or Geiger-Müller counters — all of them after a variety of radiochemical preparation, separation and extraction methods were applied.

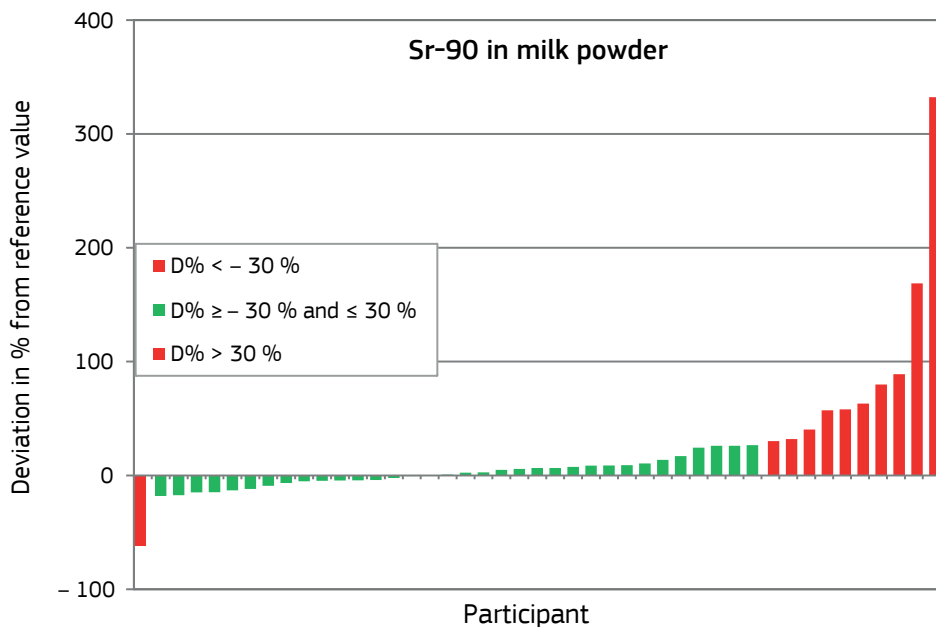
Key findings

There was a distinct difference between measurements based on GS (caesium-137 and potassium-40) and radiochemical methods (strontium-90), with the latter proving far more challenging.

The low response rate for strontium-90 was probably a consequence of the time-consuming nature of radiochemistry combined with one out of the variety of measurement methods.

Fifty-nine laboratories participated. All reported results for caesium-137 and potassium-40, but only 45 reported results for strontium-90.

Figure 3. Strontium-90 requires radiochemistry work to precede the measurements and consequently not all labs could report. The reported results were quite good with a group of labs tending to overestimate.



Notes

The reference material was obtained in the form of hard lumps that were reprocessed at JRC-Geel's laboratory for reference materials to form free-flowing powder.

Many laboratories normally monitor milk in its liquid form, and some also perform the measurements on liquid samples. In these cases the inability to reconstitute a homogeneous liquid from the milk powder may have been a possible source of error during analysis.

The full report on this PT can be downloaded free of charge from:

<https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Proficiency test III

Mineral water (2008)

Significant deviations between Member States' labs

Why was it performed?

This test was organised in anticipation of new EU requirements (in the form of a Council directive) for monitoring radioactivity in drinking water. The radionuclides below were measured in three different commercial brands of mineral water.

Radionuclides, radiation and measurement techniques

Radium-226 (alpha emitter; half-life 1 600 years); radium-228 (beta emitter; half-life 5.8 years); uranium-234 (alpha emitter; half-life 245 500 years); uranium-238 (alpha emitter; half-life 4.5 billion years).

The alpha emitters were measured using alpha-particle spectrometry (AS) (preceded by radiochemical separation). Radium-228 was measured using GS but also using LSC and proportional counters. The two latter techniques required a radiochemical separation prior to the measurement.

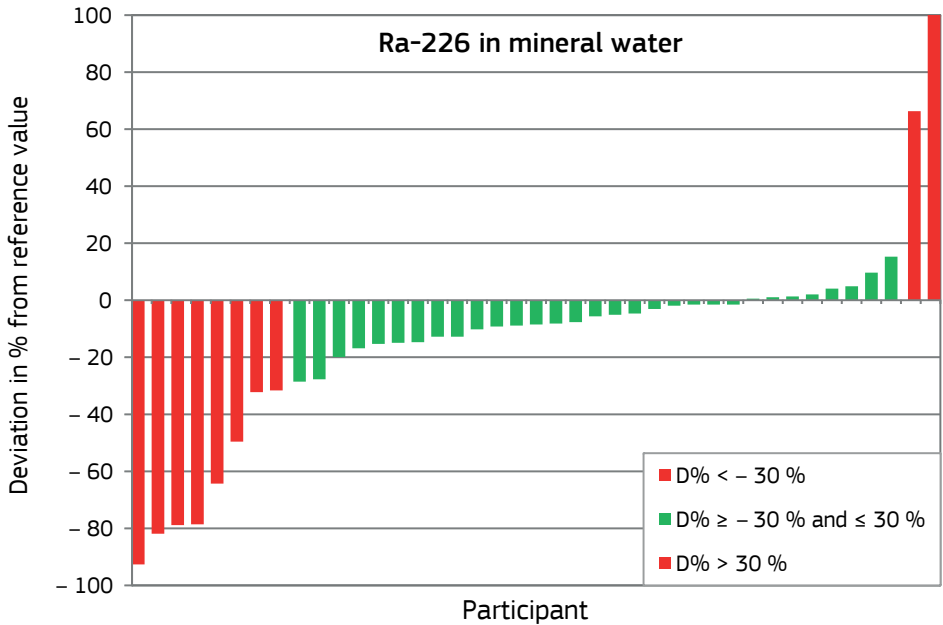
Key findings

This PT gave many unsatisfactory results. It was noted that several different methods were used by the participants.

Regarding radium, the number of discrepant measurements was alarmingly high: among the labs, about 25 % with regard to radium-226 and more than 40 % with regard to radium-228 did not pass the evaluation criteria. In 14 % of cases the results were off by more than a factor of 2.

Regarding uranium, the results were somewhat better than for radium: 6 % were off by a factor of 2 or more.

Figure 4. For radium-226, most Member States' labs produced an acceptable value, but a majority of labs underestimated the activity.



Notes

These samples had rather low activity concentrations — around the detection limits required by the draft Commission directive. Apparently, the measurement of low levels of activity is troublesome for many laboratories.

Not all laboratories are routinely analysing water for these radionuclides yet. Accordingly, the unsatisfactory comparison results for radium-226 and radium-228 were not so unexpected.

The comparison clearly demonstrates that a number of monitoring laboratories need to improve their analysis procedures for radium.

The full report on this PT can be downloaded free of charge from:

<https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Proficiency test IV

Soil (2010)

Measuring 15 radionuclides put the Member States' labs to the test

Why was it performed?

Soil is one of the most complicated environmental matrices to measure due to its complex chemistry, inhomogeneous nature and as it naturally contains many radionuclides. It is also one of the most important matrices to measure for obvious reasons, particularly following a nuclear accident. Until this exercise was carried out there had never been a soil material in the PTs in support of Article 35.

This PT put the Member States' labs to the test on how to measure a wide range of radionuclides in soil — no fewer than 15 different ones. Here, a certified reference material produced by the IAEA (No 375, Soil) was used as a base material for the comparison samples. This was of course not known to the participants.

Radionuclides, radiation and measurement techniques

^{40}K , ^{90}Sr , ^{137}Cs , ^{212}Pb , ^{212}Bi , ^{214}Pb , ^{214}Bi , ^{226}Ra , ^{230}Th , ^{232}Th , ^{234}U , ^{235}U , ^{238}U , ^{238}Pu and $^{239+240}\text{Pu}$.

To measure all of these radionuclides the Member States' laboratories needed the capacity for both GS and AS, combined with radiochemical work.

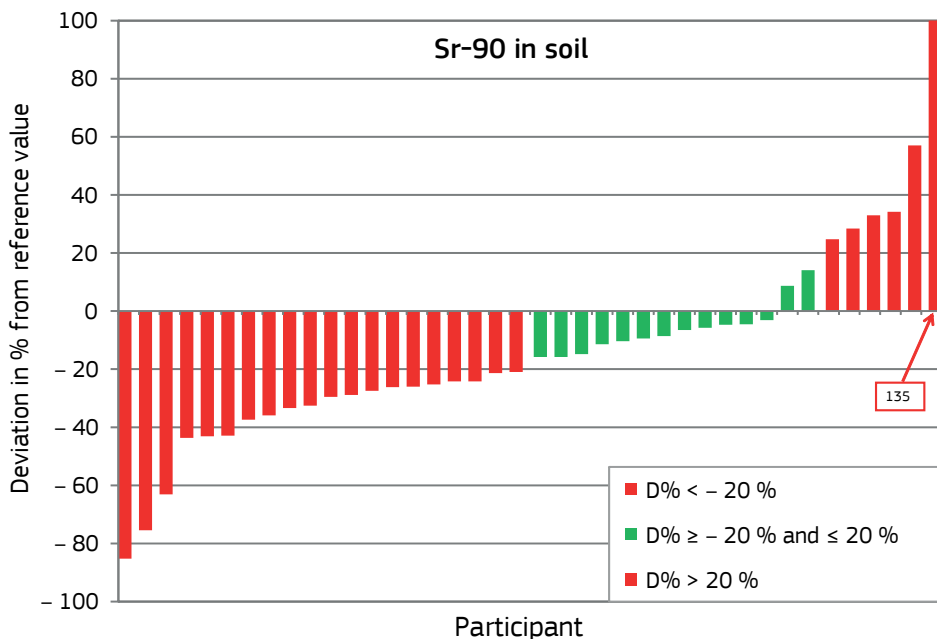
Key findings

Of 73 reporting laboratories only nine determined the activity concentrations of all radionuclides. In total, 743 results were reported, including some values below the detection limit.

Potassium-40 and caesium-137 are the radionuclides most often determined in environmental samples such as soil or sediments. The determination of these radionuclides was the least problematic for the PT participants as it is done using GS, which does not require radiochemistry.

The determination of strontium-90, which is also frequently determined in environmental samples, caused difficulties for the majority of the laboratories as it requires radiochemistry. In more than 65 % of cases the reported results deviated more than 20 % from the reference value. Accordingly, the laboratories with poor results were urged to review their analytical procedures.

Figure 5. Soil is a difficult matrix to dissolve, and consequently the strontium-90 results were not as good as, for example, those in the 2005 PT on milk powder.



Notes

The results for the determination of uranium isotopes also clearly demonstrated that several laboratories needed to improve their analytical procedures. The results for uranium-235 were found to be highly method dependent. GS rendered very poor results in comparison to AS. This is most probably due to the lack of application of appropriate interference corrections in these measurements.

A similar situation of unsatisfactory scores for GS results was observed for radium-226, and this was probably for reasons similar to those in the case of uranium-235.

One conclusion is that since no radiochemistry is needed for GS it is also used for analysing radionuclides where other methods (that do require radiochemistry) give better results.

The full report on this PT can be downloaded free of charge from:

<https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Proficiency test V

Dried bilberries (2011)

A reference material that is representative of a whole range of food products

Why was it performed?

Bilberries (European blueberries, genus *Vaccinium*) are a suitable matrix that can represent a great number of other berries and fruit. Contaminated bilberries were collected in the region affected by the Chernobyl accident, then homogenised and bottled at JRC-Geel. The reference values traceable to SI units were established in a comparison in which nine expert laboratories from national metrology institutes and the IAEA participated.

Radionuclides, radiation and measurement techniques

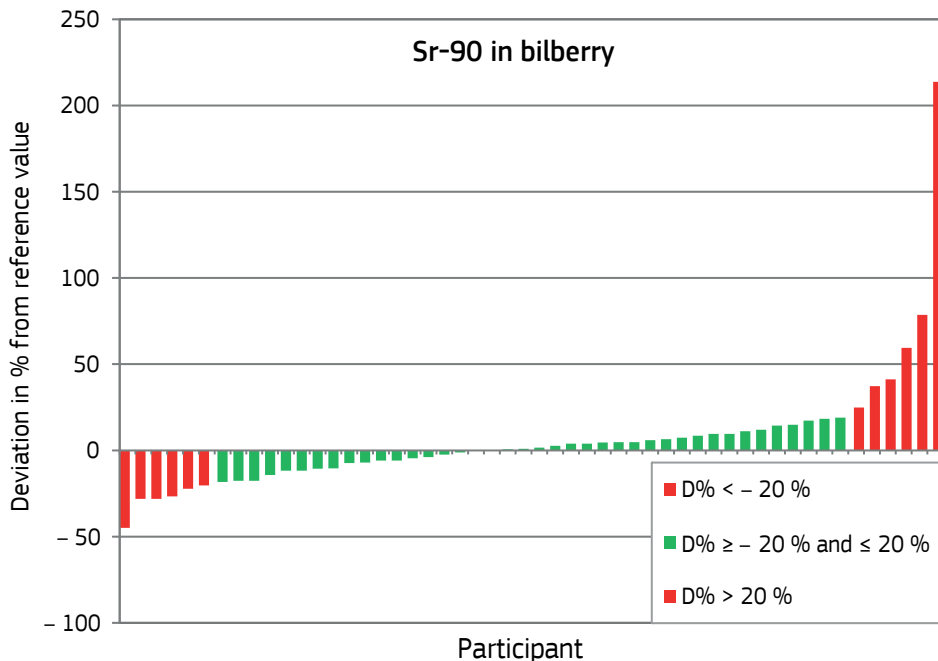
Caesium-137, potassium-40, strontium-90 (see PT II).

Key findings

The comparison demonstrates that several laboratories had difficulties in determining activity concentrations of caesium-137 and potassium-40 in food samples: in 9 % and 17 % of the labs respectively, results deviated more than 20 % from the reference values.

For strontium-90, 23 % of the labs were outside the 20 % criterion. The improved performance in comparison with the previous PTs can be attributed to the fact that it is easier to chemically separate strontium from a dried-fruit matrix than from milk powder or soil.

Figure 6. Strontium-90 is important from a radioprotection point of view as it is a 'bone seeker'. The results are quite good, and much better than for the previous PT on soil.



Notes

The results for strontium-90 are better than for the previous PT on soil. This is an indication that it is easier to perform chemical separation in plant material than in soil.

Forty-nine out of 88 participating labs submitted results for all three radionuclides.

Four labs only submitted results for one radionuclide.

Fifty-one labs submitted results for strontium-90.

Eighty-four labs submitted results for potassium-40.

Eighty-six labs submitted results for caesium-137.

The full report on this PT can be downloaded free of charge from:

<https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Proficiency test VI

Mineral water (2012)

Highlighting the diversity of lab practices and a controversial analytical method: gross counting

Why was it performed?

This PT was conducted by 71 participating labs in anticipation and support of the new Euratom drinking water directive (Council Directive 2013/51/Euratom), which includes gross alpha and beta activity screening levels.

Radionuclides, radiation and measurement techniques

No specific radionuclides were measured in this PT. Instead, gross counting methods were used where the gross alpha or beta activity is measured. Three different water reference materials were provided to each lab. Each lab could thus report six values.

Key findings

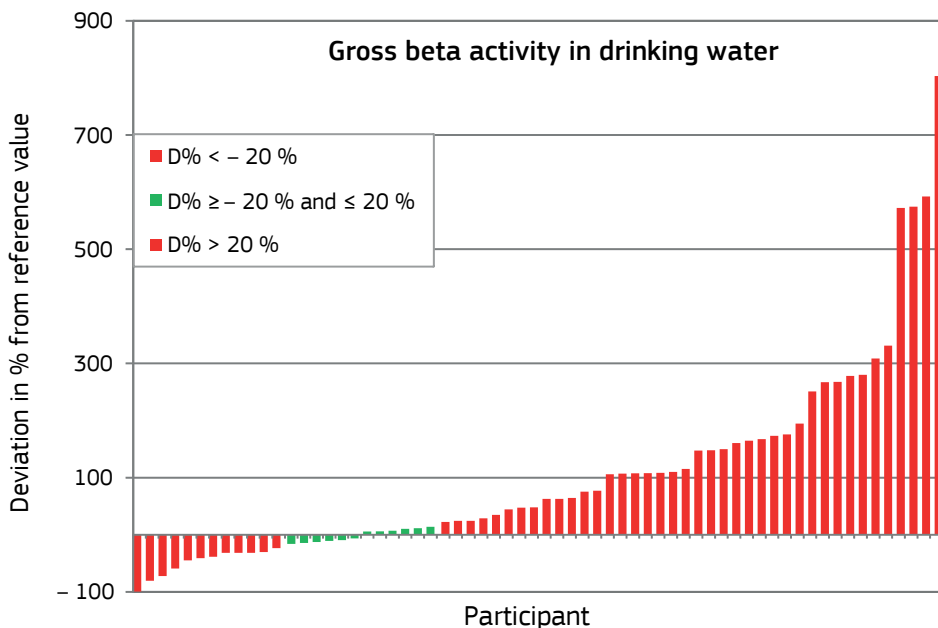
Only one laboratory was able to determine gross alpha/beta activities within the reference range ($\pm 30\%$ from the reference values) for all three waters. Furthermore, only 42 % of the participants could report at least half of the six results within the reference range.

In total more than half of the laboratories had severe problems. For example eight laboratories (11 %) were not able to report any measurement data within the reference range at all.

None of the gross counting methods used by participants proved to be superior to the others. Even the application of the same method in different laboratories did not guarantee comparable results.

Gross counting methods can be treacherous, as different radionuclides give different responses in the detectors used. The detector response can be highly unpredictable if a sample contains radionuclides that are different from those used when calibrating an instrument.

Figure 7. A typical graph showing the results from gross counting methods on mineral water. The spread of results is very wide, which shows that there is not sufficient metrological control in the majority of labs.



Notes

Some of the reasons for the unsatisfactory results were as follows.

- The initial radionuclide composition of the samples was not known beforehand, and the gross counting technique is sensitive to changes in the radionuclide composition compared to the calibration samples.
- One of the samples had a very low gross alpha activity concentration.
- Sample preparation methods (possible loss of volatile radionuclides).
- The time delay between sample preparation and measurement (ingrowth of radon and its progenies).
- Method-specific pitfalls: since ^{40}K is not co-precipitated it is excluded from the gross beta results, whereas if direct evaporation or LSC is used then ^{40}K contributes to the gross beta activity.

The full report on this PT can be downloaded free of charge from:

<https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Proficiency test VII

Air filter (2014)

The 2003 exercise repeated — with more labs and similar results

Why was it performed?

This exercise was in principle identical to the exercise in 2003 and could therefore be used as a good benchmark on possible improvements in laboratories' measurement results. Just like in 2003, the participants sent their own filters to JRC-Geel where they were spiked with caesium-137. Of 76 participating laboratories in 2014, 30 had participated in the 2003 exercise.

Monitoring radioactivity in air is of paramount importance. There are about 5 000 stations in Europe for monitoring dose rates (with no information on the radionuclide), but 'only' around 250 stations for sampling particulates in air on filters, from which quantitative radionuclide information can be obtained.

Radionuclides, radiation and measurement techniques

See PT I.

Key findings

Only five laboratories reported results that deviated more than $\pm 33\%$ from the reference value. A small improvement of acceptable relative deviation (from 87 % to 93 % of the labs) was seen compared to the PT in 2003.

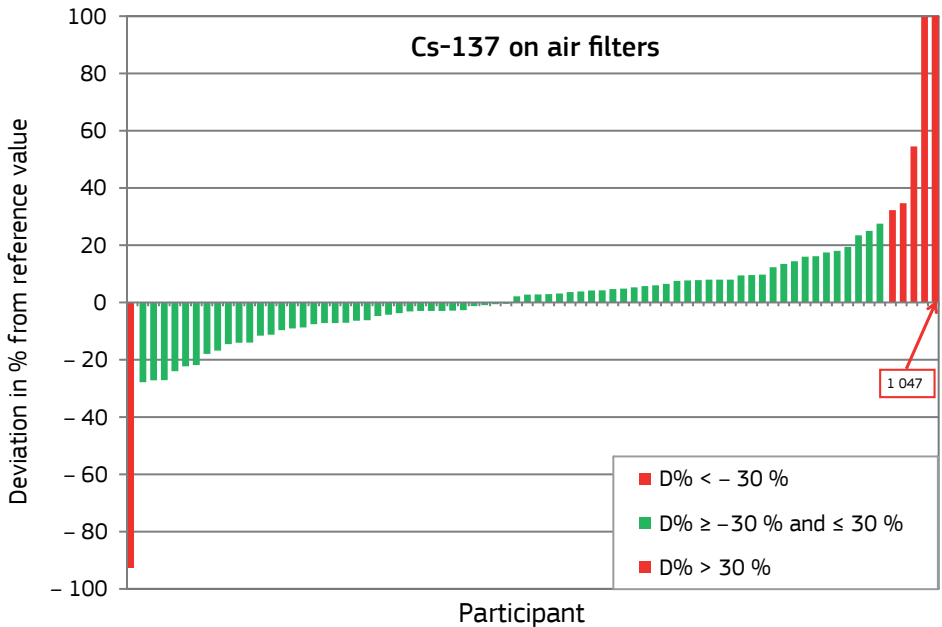
When including the uncertainty estimation in the evaluation, 23 laboratories reported results that were not compliant.

The types and sizes of filters used in Member States' labs are highly diverse. For this PT they ranged in size from 17 cm² to 4 200 cm². The most common types are made of either nitrocellulose, polypropylene or glass fibres. No correlation was found between the filter used and the relative deviation. However, the labs with large filters obtained higher E_n numbers (underestimation of uncertainty).

There is no harmonised best practice. The amount of air that is sampled varies greatly, as do the sampling intervals (daily, weekly, monthly, annually or ad hoc).

Of the 30 laboratories that participated in 2003, nine improved their results. However, seven labs that reported acceptable results in 2003 are now non-compliant (with regard to either E_n number or relative deviation).

Figure 8. Air filters constitute a very simple matrix and this result from caesium-137 confirms the good standard of the vast majority of the labs.



Notes

The 33 % range (also used in the 2003 exercise) was chosen, taking into account the low caesium-137 activity level and the non-homogeneous distribution of the activity on the air filters. The activity on each filter varied — as a consequence of the different types of filters the participants use — from 70 mBq to 2.3 Bq (a factor of 32).

Only one lab used a NaI detector (with poor resolution) and the remaining 75 labs used an HPGe detector (with high resolution).

The full report on this PT can be downloaded free of charge from:

<https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Proficiency test VIII

Air filter (2016)

Highlighting problems with Cs-134 and I-131

Why was it performed?

Formally, this exercise was conducted as part of a European metrological research project funded by Euramet. For this reason, the identity of each lab was not revealed to either the Commission's Directorate-General for Energy or to the national regulatory authority. However, the PT was similar to the exercises conducted in 2003 and 2014, with the difference that caesium-134 and iodine-131 were also included. Out of the 67 participants, 57 (85 %) also participated in 2014. In total, 26 laboratories participated in all three air-filter PTs.

Radionuclides, radiation and measurement techniques

Caesium-137 (see PT 1).

Caesium-134 (beta decaying but also emitting gamma rays; half-life 2 years).

Iodine-131 (beta decaying but also emitting gamma rays; half-life 8 days).

All three radionuclides are very important to monitor following a nuclear accident using GS.

The results were such that 56 out of the 67 participating laboratories (84 %) reported values within the ± 20 % range of the reference value for both caesium-137 and caesium-134, but for iodine-131 it was only 20 (30 %) laboratories. The reasons for the reported results generally being too low for iodine-131 are as follows.

- The filters from different countries are made from different materials. Some materials are better than others in retaining the iodine.
- Some laboratories use other types of filters for measuring iodine in air than those used in this PT (e.g. using trapping in activated carbon).
- Although all participants were instructed to keep the filter inside the plastic bags in which they were delivered, some laboratories took out the filter before the measurements, which caused a loss of iodine.
- Some laboratories admitted to drying the filter (as instructed in their own written procedures), which of course led to a loss of iodine.
- Failing to perform a correction for decay during measurement or a normal decay correction can result in too low a result for iodine-131, which has a short half-life (8 days).

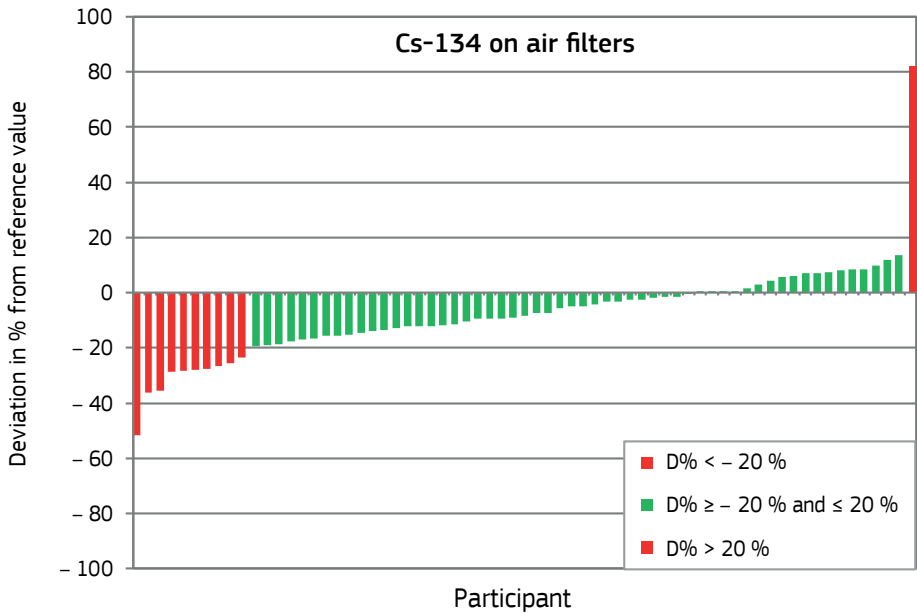
Key findings

For caesium-134 many laboratories reported too low a value. The key reason for this is a failure to perform a coincidence-summing correction.

When comparing caesium-137 with the PTs in 2003 and 2014, one can observe a slight improvement over the years.

The PT highlights the need of further study to better understand the discrepancies for iodine-131.

Figure 9. Results from caesium-134 measurements often show a slight negative bias, which is also the case here.



Notes

Iodine-131 has a short half-life (8 days) and is therefore of great concern during the first weeks following a nuclear accident, particularly as it induces thyroid cancer, predominantly amongst children. Iodine is also a volatile element that is easily lost from a sample, and is therefore of concern as the level can easily be underestimated.

The full report on this PT can be downloaded free of charge from:

<https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Proficiency test IX

Dried maize (2017)

Emergency reporting introduced for the first time

Why was it performed?

The aims of this PT were:

- to verify the performance of Member States' labs on a feed matrix (maize) and the three most important radionuclides following a nuclear accident;
- to verify the performance of the Member States' labs in performing rapid measurements and reporting as requested in case of a real accident scenario.

This was the first time in the JRC's PT scheme for radioactivity that emergency reporting was introduced. The labs were asked on a voluntary basis to submit their results within 48 hours from the receipt of the sample. For the 'normal' compulsory (routine) reporting the labs had about 2 months.

Radionuclides, radiation and measurement techniques

Caesium-134, caesium-137, iodine-131 (see PT VIII). All three radionuclides are easy to measure using GS with an HPGe detector.

The maize powder was produced from commercially available maize grains intended for usage as feed. The production went through the following steps: cryomilling, sieving, milling, sieving, mixing, spiking with radioactivity, mixing, milling, mixing and, finally, bottling.

Key findings

There were 120 laboratories that submitted results within the 2-month deadline.

Seventy laboratories (more than half of the total number of participants) submitted results for the (voluntary) emergency reporting.

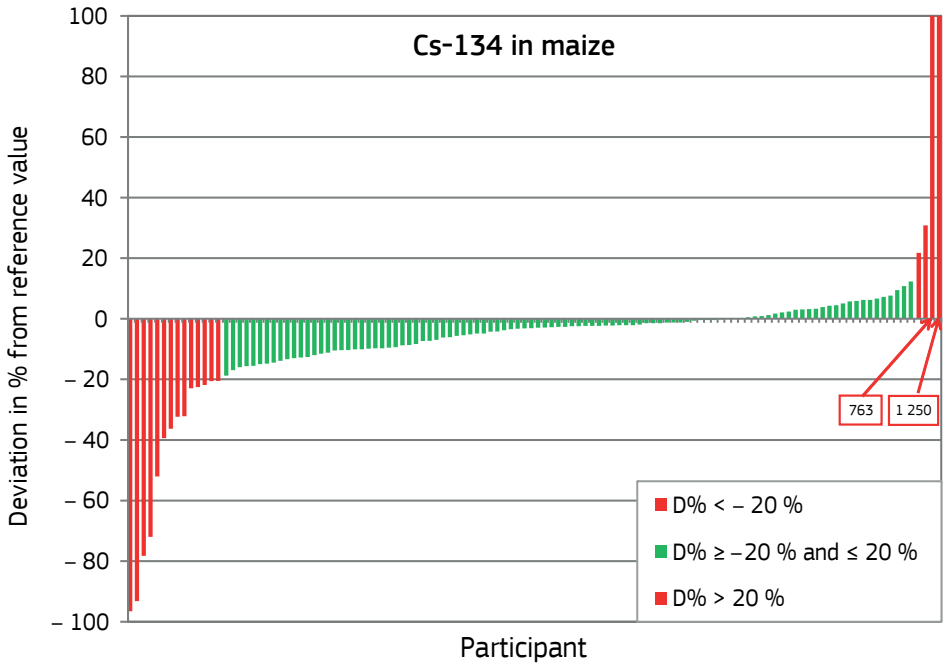
The overall results were better for the emergency reporting than for the routine reporting, indicating that the laboratories that participated in the emergency exercise are probably the more experienced/competent ones.

The results for caesium-137 and iodine-131 showed no particular bias, i.e. about the same number of labs measured values that are slightly too low as measured values that are slightly too high.

As in all previous PTs there were many labs that measured values for caesium-134 that were slightly too low. This was most likely due to not performing coincidence-summing correction, which is important for caesium-134.

Except for a few outliers, the general measurement precision was good for all three radionuclides.

Figure 10. As usual, many labs measure caesium-134 values that are slightly too low (due to coincidence summing), but most labs do a good job.



The full report on this PT can be downloaded free of charge from:

<https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Proficiency test X

Drinking water (2018)

Observation: loss of the inert gas radon causes underestimation of activity

Why was it performed?

Radon is a naturally occurring radioactive gas that is very bad to inhale due to the alpha particles that it emits. It has also many radioactive progenies, and in fact 1 Bq of radon-222 will result in 8 Bq of total activity if the progenies are contained. Due to the associated health hazards radon is heavily regulated. In 2013 the Council issued a new directive, the Euratom drinking water directive. This PT was organised as a response to requests from Member States' experts and DG Energy to verify drinking-water controls.

In addition, due to the short half-life (3.8 days) and problems of sampling and storage there have been very few intercomparisons or PTs in the past. There is therefore a need for harmonisation activities when it comes to radon-222.

Radionuclides, radiation and measurement techniques

Radon-222 (alpha decaying; half-life 3.8 days).

Three methods were used by the participants: (i) GS of the radon progenies, (ii) LSC and (iii) emanometry.

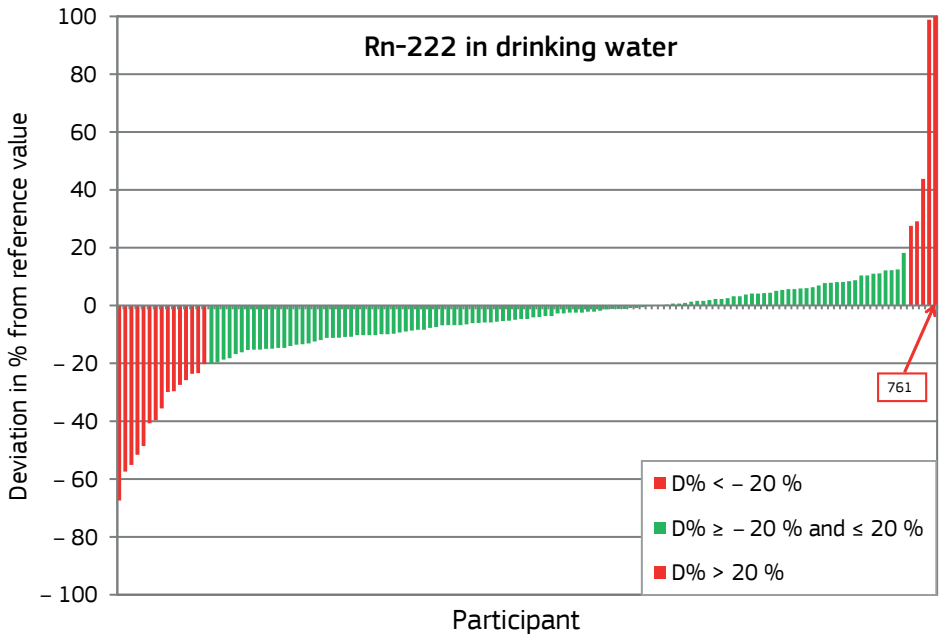
Key findings

We found that there were numerous sources (see notes on the following page) leading to radon loss that would generate a measurement result that is too low (see graph) — something that is problematic from a radioprotection perspective.

The measurement methods used all seem to be fit for purpose, and it cannot be concluded that one method did better compared to another.

However, the reproducibility of standardised techniques was much better than that from non-standardised techniques.

Figure 11. The results with a slight negative bias indicating loss of radon.



Notes

The Euratom drinking water directive is formally called [Council Directive 2013/51/Euratom](#) of 22 October 2013 laying down requirements for the protection of the health of the general public with regard to radioactive substances in water intended for human consumption.

Radon-222 is an inert gas (does not react chemically) and can therefore easily escape from the sample and measurement volume, making all methods inherently less robust if care is not taken during sampling, transport and storage.

Based on feedback from Member States' labs and experts we are seeking to organise a PT involving sampling and transport in the future (as this has never been done before). Radon in drinking water would be the obvious choice for such a PT.

The full report on this PT can be downloaded free of charge from:

<https://remon.jrc.ec.europa.eu/Services/Proficiency-Tests>

Future directions

Consequences of measurement errors

Measurements by monitoring laboratories impact both financial and health aspects of society. Reporting numbers that are **too high** may lead to unnecessary remedial and legal actions and a difficult process to remove products from the market. In addition, it may cause stress for people who think they may have been exposed to dangerous levels of radiation. Experiences from Chernobyl and Fukushima show that the latter point needs to be taken very seriously.

Reporting values that are **too low** may cause: (i) long-term health effects in workers and the general public due to the failure to detect a radiation hazard; (ii) a failure to properly trace back the source of an undisclosed release of radioactivity; (iii) possible overestimation of the impact of future releases due to having values at present that are too low.

If Chernobyl happened today

In comparison to 1986 the monitoring labs in the EU are now much better prepared to handle a situation like Chernobyl. Measurement equipment has improved and the awareness of what samples to measure and which radionuclides to look for is greater. Many labs also conduct regular emergency exercises.

There are, however, some things to consider. During emergency situations other methods are used compared to routine monitoring. In addition, staff may have to cope with contaminated labs and stress. In the future Member States may have to look further into such aspects, which are more cumbersome to test.

A list of future actions

Knowledge

The most important action is to make sure that there are sufficient people with the right education to operate the labs in the correct way. In particular there are concerns regarding the number of radiochemists being trained in Europe today.

Underperforming labs

In every PT there are a few labs that underperform and report data far from the reference values. It is important that Member States take action to help these labs improve. In some cases such issues may be due to an occasional error, but in many cases they are more structural in nature. Again, training and PT participation offer important pieces of the cure.

Sampling and measurements

The results reported in our PTs can be considered a best-case scenario, where the labs measure a well-characterised matrix. In a real-world scenario they will most likely face challenges during sampling and transport. Work is needed in the future on how to ensure representative samples, and for example how to handle the volatility of substances such as iodine or inert radon.

In some cases the measurement methodology needs to be developed. Some methods, such as gross counting methods, should be used with great caution, or even phased out altogether, highlighting the need to replace the method of choice at the laboratory with more robust methods.

Reference labs

The Fukushima accident demonstrated the important role of reference labs and how they could help less-experienced labs improve. The staff in such labs should also lead work on international (written) standards — work that is crucial for monitoring activities, but difficult as it requires funding from the ‘home institute’ (and not from the standardisation body). Furthermore, the development of reference materials is essential: the present stock of reference materials is decaying, and new materials and radionuclides need to be added.

Neighbouring countries

From an EU perspective, it is reassuring when fellow Member States cooperate to implement a solid monitoring capacity across Europe. Not only does this help the EU broadcast an early warning, but it also guarantees that a minimal amount of radioactive products will spread from a contaminated region.

Science

Finally, we can also see that monitoring labs can contribute to scientific studies. By establishing baseline radioactivity levels (i.e. the present level of radioactivity in the environment), it becomes easier to assess the impact of future anthropogenic activities. Radioecological data are also needed to establish the uptake of various radionuclides in different organisms and the impact of different soil conditions. Such research will help to assess the safety of agriculture on slightly contaminated land with an eye to establishing whether the final product is good for consumption.

ANNEX

Introduction to radioactivity
measurement and the activities
at JRC-GEEL — A brief tutorial

Measuring radioactivity

Normally, three types of radiation are measured

Radioactivity is the process by which the nucleus of an unstable atom changes, and emits ionising radiation. The most well-known types of ionising radiation can be seen in the table below.

Table A1. *Brief description of three types of ionising radiation.*

Type of radiation	Description of the radiation	Penetrative power
Alpha particles	Helium-4 nucleus (2 protons + 2 neutrons)	Stopped by paper; cannot penetrate skin
Beta particles	Electrons	Stopped by a sheet of metal
Gamma rays	Electromagnetic radiation	Attenuated by heavy elements such as lead

In total there are around 3 500 radionuclides

On earth around 95 naturally occurring elements are present. They can exist in thousands of different varieties depending on the configuration of their nucleus. About 3 500 of these varieties have been identified as being radioactive; these are what are referred to as **radionuclides**. They disintegrate while emitting radiation — some very quickly, some very slowly.

For direct societal needs it is important to monitor or study around 200 radionuclides on a regular basis. This includes both naturally occurring radionuclides and man-made radionuclides used in medicine and industry. However, all 3 500 are interesting for science.

The becquerel unit

The activity of a radioactive material is measured using the becquerel (Bq) SI unit. This unit represents the number of atoms that decay (disintegrate) in 1 second. The BIPM, which is located in Paris, is in charge of defining and realising all SI units, including the Bq.

The half-life indicates the rate of decay

The half-life is a concept that is used to describe how quickly a radionuclide decays. It is defined as the time required for 50 % of the atoms to disintegrate. A substance with a short half-life will thus quickly become harmless to humans. For example iodine-131 loses half of its activity in 8 days.

Table A2. *'The usual suspects' — a few examples of radionuclides that are important to monitor.*

Radionuclide	Half-life	Decay mode	Typical source
Potassium-40	1.3 billion years	Beta decay (*)	Natural
Caesium-134	2 years	Beta decay (*)	Nuclear accident
Caesium-137	30 years	Beta decay (*)	Nuclear accident
Strontium-90	29 years	Beta decay (*)	Nuclear accident
Iodine-131	8 days	Beta decay (*)	Nuclear accident, release from hospitals, etc.
Radium-226	1 600 years	Alpha decay (*)	Natural
Radium-228	6 years	Beta decay (*)	Natural
Thorium-232	14 billion years	Alpha decay	Natural
Radon-222	3.8 days	Alpha decay	Natural
Uranium-238	4.5 billion years	Alpha decay	Natural (also in nuclear fuel)
Hydrogen-3; Tritium	12 years	Beta decay	Release from nuclear facility

(*) Accompanied by a subsequent strong gamma-ray emission.

Why we measure radioactivity

Radiation is naturally present everywhere

Radioactivity is a naturally occurring phenomenon. Here on earth it can be detected everywhere: in water, soil, rock, living organisms such as plants and animals, construction materials, etc. There is, for example, around 100 Bq/kg naturally present in the human body (mainly potassium-40 and carbon-14, the latter commonly used for the age estimation of ancient organic material).

Ionising radiation can be harmful in two ways

Although ionising radiation damages DNA and cells, it is not dangerous at the 'normal' levels present in and around us as the body's natural repair mechanisms are tailored to handling this. At increased levels, however, ionising radiation is dangerous. The harmful effects are often divided in two categories.

- **Deterministic effects.** Direct damage to tissues as a result of the death or malfunction of cells. This includes acute radiation sickness and burns that appear shortly after irradiation.
- **Stochastic effects.** Damage to DNA, leading to random mutations that may or may not prove harmful over a longer course of time, in some cases causing cancer or hereditary defects.

Inside/outside the body

Different radionuclides affect the body differently depending on the radiation they emit and which organ is being targeted.

Alpha emitters deposit a large amount of energy within a short range and are therefore more dangerous when they are inside the body than outside. An example is the radioactive noble gas radon, which together with its decay products causes great damage to the lungs when inhaled.

Beta emitters can also cause problems inside the body. An example is iodine-131, which accumulates in the thyroid gland and may cause local damage.

To describe the rate at which different radionuclides enter and leave the body the term biological half-life is often used. Caesium-137, for example, is removed from the body relatively fast. Its biological half-life is about 70 days, although its physical half-life is 30 years. In contrast some radionuclides are said to be 'bone seekers', and stay in the skeleton for a very long time. An example of this is strontium-90, for which the (highly uncertain) biological half-life is between 18 years and 50 years.

Harmful radiation can come from different sources

Increased levels come mostly from the following sources.

- **Major nuclear accidents.** For example the wind-borne particles from the Chernobyl accident in 1986.
- **Accidental releases from nuclear installations.** Several leaks of gaseous iodine-131 from facilities producing or using medical radioisotopes have been reported.
- **Hospitals and industry.** For example obsolete industrial or medical equipment containing cobalt-60.
- **Natural sources.** For example radon-222 from the decay of radium and uranium in the ground and in building materials.

Case studies

Below are some cases that illustrate the ever-present need for radioactivity monitoring. More case studies can be found on Wikipedia ⁽¹⁾ or on the IAEA website ⁽²⁾.

Chernobyl 1986

The most well-known nuclear accident took place in the Soviet Union on 25 and 26 April 1986. A nuclear plant close to the town Pripyat (in present-day Ukraine) suffered an explosion in one of its reactors. Consequently, radioactive material was spread all over Europe on easterly winds. Although there was no official reporting about the accident, radioactive contamination was monitored by a number of European states. The first observations were made on 28 April at Forsmark nuclear power plant on the east coast of Sweden, where the staff were found to have radioactive particles on their workwear.

Radioactive metal being handled as scrap

Sometimes batches of radioactive metals go astray, typically when radiation equipment is scrapped in countries with a poor monitoring structure. A common example is cobalt-60, which is widely used for medical applications, not least during external beam radiotherapy for cancer.

Radioactive metals may look to the eye like any harmless scrap metal. Over the years this has led to a number of accidents in different places. In some extreme cases radioactive scrap metal has even been melted and recycled. As late as 2013 the metal studs of a leather belt sold by an online retailer tested positive for cobalt-60; this was monitored by US border control and led to a worldwide recall of the product.

Another illustrative incident took place in Mexico in 2013. Robbers stole a lorry carrying cobalt-60 from obsolete radiation therapy equipment that was on its way to a waste facility. One of the robbers was subjected to radiation as he removed the material from its protective casing and spilled the cobalt-60 pellets on the ground.

⁽¹⁾ https://en.wikipedia.org/wiki/List_of_civilian_radiation_accidents

⁽²⁾ https://web.archive.org/web/20170503063725/http://www-pub.iaea.org/books/IAEABooks/Publications_on_Accident_Response; <https://www-news.iaea.org/EventList.aspx?ps=100>

Fukushima 2011

On 11 March 2011 an earthquake of moment magnitude 9.1 off the east coast of Japan caused a gigantic tsunami. It destroyed a number of villages along the coast and killed almost 20 000 people.

The nuclear power plant at Fukushima was also hit but at first seemed to suffer no serious damage as the shutdown worked well. Unfortunately, the reserve generators responsible for driving the cooling of the very hot reactor fuel were damaged. This led to the overheating of the fuel and subsequent chemical explosions in which radioactivity was released into the air and sea. Japan quickly realised that their monitoring capacity was not sufficient to guarantee the safe use of food products. Many new laboratories were therefore quickly set up with the task of checking food (and other) products. To guarantee the quality of the results from these new laboratories several Japanese expert labs produced new reference materials (rice, fish, etc.) for these labs and started organising PTs. Studies have shown that when the public has the possibility to have their own home-grown food products checked for radioactivity, the psychological effect is very positive and greatly influences the general well-being of the public.

Undisclosed accidents

With the improvement of monitoring networks it is more difficult for operators to get away with undisclosed events. Below is a recent example.

In early October 2017 many European labs started detecting ruthenium-106 in filters they used for monitoring radioactivity in air. This is an unusual radionuclide to detect in air, particularly when it is detected by itself and is not accompanied by other radionuclides. The activities were very low and posed no danger to human health in the areas in the EU where it was detected. Still, it was a mystery that it had showed up. Ruthenium-106 is used for the treatment of cancer of the eye, but it was unlikely that the release could come from such production.

By collating data from European laboratories and comparing them with meteorological data it was possible for researchers to claim with high probability that the release came from a specific region and was probably in the order of more than 100 TBq (100 trillion Bq) ⁽³⁾.

⁽³⁾ https://www.irsn.fr/EN/newsroom/News/Documents/IRSN_Information-Report_Ruthenium-106-in-europe_20171109.pdf
<https://doi.org/10.1073/pnas.1907571116>

Activities at JRC-Geel

The Member States of the European Union continuously monitor radioactivity in the environment

Chapter 3 of the Euratom Treaty (Articles 30-39) specifies what actions the EU Member States must take to guarantee public safety. Accordingly, Article 35 obliges the EU Member States to monitor radioactivity in the environment. This work involves the following two responsibilities.

- The regular sampling and measurement of various specimens (air, water, soil, food feed, etc.) to confirm that there are no alarming deviations from base values.
- The preparedness for emergency scenarios when it may be necessary to process vast numbers of samples within a short period of time.

The JRC makes sure these monitoring data are accurate and available

The Euratom Treaty also obliges the Commission to verify that these operations are performed up to standard (Articles 35 and 36). Furthermore, it states that the JRC shall assist the Commission in these tasks (Article 39). The following JRC services carry out these assignments:

- JRC-Geel (northern Belgium; called IRMM until 2016) verifies the quality of the monitoring results by organising PTs.
- JRC-Ispra (northern Italy) collects and publishes the monitoring data that the Member States' laboratories report.

JRC-Geel represents a stamp of quality

The laboratories at JRC-Geel bring together some of Europe's leading experts and represent the most advanced instrument park for radioactivity measurement. As they specialise in developing and performing highly accurate measurements and preparing reference materials, their activities can be seen as a benchmark and a stamp of quality.

JRC-Geel gives valuable feedback to the labs of the Member States and contributes to ensuring the following.

- No costly or harmful decisions are taken from faulty data.
- EU citizens can trust the information and recommendations from their authorities.

In addition, JRC-Geel helps the EU and the Member States to trace sources of unreported radioactivity release.

The collated data also form a useful resource for scientists

Each year, more and more members of the scientific community realise that the freely obtainable data from the JRC and the Member States can be used in areas far beyond nuclear physics, for example:

- to understand the climate and the environment;
- to validate computer models that predict, for example, atmospheric dispersion.

JRC-Geel's core competence: proficiency tests

Proficiency tests confirm the skill of Member States' monitoring labs

Quis custodiet ipsos custodes? — 'Who watches the watchmen?' This classic Latin phrase points out why PTs are needed and performed.

In a PT the organiser distributes the same reference material to all participating labs. The labs measure what is asked (in this case the unknown activity of specific radionuclides) and report back to the organiser. The organiser summarises the results and informs the laboratories of whether they passed or failed (or provided questionable results).

All results are publicly available, but it is not revealed which result belongs to which laboratory. However, for each Member State, the national regulatory authority receives detailed information on how that country's labs are performing.

The PTs are requested by DG Energy based on input from the Member States

The PTs are initiated by the Commission's DG Energy. DG Energy organises meetings on a regular basis with Member States' experts, at which issues connected to Articles 35 and 36 of the Euratom Treaty are discussed. During these gatherings the participating experts (representing the EU Member States) can express their wishes for matrices and radionuclides they consider to be of particular importance. After feedback from JRC-Geel a decision is normally taken at the next meeting.

Producing a reference material requires special skills

JRC-Geel specialises in producing large quantities of reference materials on an industrial scale in its reference material laboratory. It is essential that the reference material be of the highest quality (stable and homogeneous) and that the reference values be undisputed, robust and set before the exercise, just like the evaluation criteria. Producing a good-quality reference material of a 'natural matrix' can be a complex procedure, involving highly controlled sampling, cutting, crushing, mixing, sieving, etc.

There are two categories of reference materials.

- Spiked materials, for which the radioactivity is added in the laboratory.
- Natural radioactive materials, for which the radioactivity is taken up by the organism (plant/animal) or incorporated into the matrix (soil, sediments).

Reference values are determined by spiking or reference measurements

Once the homogeneity of the samples is established, reference values are determined. This can be done in different ways, depending on the reference material and radionuclides.

- **Spiking a standardised solution**, i.e. adding a known amount of radioactivity to a water-based solution (e.g. mineral water).
- **Spiking a solid material**. Usually a slurry is spiked and then dried. This is a more complicated action that requires extensive experience and quality checks.
- **Reference measurements of a sampled material** (e.g. grass from the Chernobyl area). This is the alternative which is closest to a real-case scenario but difficult as no a priori knowledge of the activity is available. Therefore, reference measurements by the JRC and as well as trusted laboratories have to be made to establish the actual activity level.

Three main criteria are used: relative difference, z- and zeta score (previously also E_n number)

The PTs follow the recommendations of the ISO 17043 standard, which prescribes a number of different evaluation criteria. The two main evaluation criteria are as follows.

- **Relative difference** (between the reported value and the reference value). This is the most basic evaluation criteria. It is also the only one shown in graphs in this brief report.
- **Z score**. This score takes into account the difference between the reported value and the reference value and compares the difference to the standard deviation of the proficiency test.
- **Zeta score**. This is an important score as it takes into account not only the deviation in the result but also the reported uncertainty. Reporting a correct uncertainty is necessary to understand how much weight one can give to a certain measurement when taking a decision. (In earlier PTs the E_n number was used in a similar way, but it has now been replaced by the zeta score.)

A number of detection techniques are used by the Member States

Measuring radioactive decay is both a science and art in itself. The table below summarises some of the most common methods used.

Table A3. *Brief description of common radioactivity measurement techniques.*

Method	Principle	Used in PTs
Gamma-ray spectrometry (GS)	Direct measurement of gamma rays with an energy-sensitive radiation detector nowadays mostly made of high-purity germanium.	All except No. VI
Liquid scintillation counting (LSC)	The liquid sample is mixed into a cocktail of chemical substances that give away light when hit by ionising radiation. This light emission is measured.	II, III, V, VI, X
Alpha-particle spectrometry	Direct measurement of alpha particles. Nowadays mostly with a silicon semiconductor detector.	III, IV
Solid-state scintillation counting	Alpha particles produce light in the solid detector. The light is detected using photomultipliers.	II, VI, X
Gas-flow proportional counting	A detector filled with a gas (e.g. argon-methane gas mixture) inside which a high voltage is placed. Alpha and beta particles will induce electrical pulses when ionising the gas.	II, III, V, VI
Emanometry	A technique in which radioactive substances are degassed into air/gas followed by detection of radiation/particles present in the air/gas using different detection techniques (e.g. air ionisation, solid-state scintillators or semiconductors).	X

We distinguish between ‘counting’ and ‘spectrometry’.

- During **counting** one has to make sure that there is only one radionuclide in the sample and every count in the detector comes from that specific radionuclide.
- **Spectrometry** is useful when many radionuclides are present in the sample. Each radionuclide emits distinct gamma rays or alpha particles, which will give rise to peaks in a spectrum. The size of each peak is proportional to the amount (or activity) of that specific radionuclide.

List of initialisms

AS	alpha-particle spectrometry
BIPM	International Bureau of Weights and Measures
EU	European Union
GS	gamma-ray spectrometry
HPGe	high-purity germanium
IAEA	International Atomic Energy Agency
JRC	Joint Research Centre (of the European Commission)
LSC	liquid scintillation counting
PT	proficiency test
SI	International System of Units

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