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Radiation Protection: the experience of the Integrated Laboratory of Radioactivity Measurement and Monitoring of the ENEA Radiation Protection Institute

Sandro Ridone

sandro.ridone@enea.it

ENEA

Dolores Arginelli

National Agency for New Technologies, Energy and Sustainable Economic Development

Maria Chiara Botta

National Agency for New Technologies, Energy and Sustainable Economic Development

Daniela Gorietti

National Agency for New Technologies, Energy and Sustainable Economic Development

Francesco Modestia

National Agency for New Technologies, Energy and Sustainable Economic Development

Davide Pirullo

National Agency for New Technologies, Energy and Sustainable Economic Development

Salvatore Zicari

National Agency for New Technologies, Energy and Sustainable Economic Development

Paolo Battisti

National Agency for New Technologies, Energy and Sustainable Economic Development

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Name of the authors:
[1]Sandro Ridone, [1]Dolores Arginelli, [1]Maria Chiara Botta, [2]Daniela Gorietti, [2]Francesco Modestia, [2]Davide Pirullo, [3]Salvatore Zicari, [4]Paolo Battisti

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Affiliations and addresses of the authors:
[1] ENEA, Radiation Protexction Institute, Integrated Laboratory of Radioactivity Measurement and Monitoring: strada per Crescentino 41, 13040 Saluggia (VC), Italy;
[2] ENEA, Radiation Protexction Institute, Integrated Laboratory of Radioactivity Measurement and Monitoring: via Anguillarese 301, 00123 Rome, Italy;
[3]ENEA, Radiation Protexction Institute, Integrated Laboratory of Radioactivity Measurement and Monitoring: SS 106 Jonica km 419+500, 75026 Rotondella (MT), Italy;
[4]ENEA, Radiation Protexction Institute, Integrated Laboratory of Radioactivity Measurement and Monitoring:via del Mille 31, 40121 Bologna, Italy.

E-mail address of the corresponding author:
sandro.ridone@enea.it

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Sandro Ridone[1*], Dolores Arginelli[1], Maria Chiara Botta[1], D. Gorietti[2], F. Modestia[2], D. Pirullo[2], S. Zicari[3], Paolo Battisti[4]

[1] ENEA, Radiation Protexction Institute, Integrated Laboratory of Radioactivity Measurement and Monitoring: strada per Crescentino 41, 13040 Saluggia (VC), Italy;
[2] ENEA, Radiation Protexction Institute, Integrated Laboratory of Radioactivity Measurement and Monitoring: via Anguillarese 301, 00123 Rome, Italy;
[3]ENEA, Radiation Protexction Institute, Integrated Laboratory of Radioactivity Measurement and Monitoring: SS 106 Jonica km 419+500, 75026 Rotondella (MT), Italy;
[4]ENEA, Radiation Protexction Institute, Integrated Laboratory of Radioactivity Measurement and Monitoring:via del Mille 31, 40121 Bologna, Italy

Abstract

Since many years ENEA Integrated Laboratory of Radioactivity Measurement and Monitoring has been working in the field of radioactivity measurement especially for environmental and internal dosimetry monitoring purposes in the Centers of Saluggia, Casaccia and Trisaia. The large set of measurement systems and equipment, with the knowhow for the application of specific techniques of physical-chemical and radiochemical preparation, allow to carry out analyses of different samples for the determination of a huge number of radionuclides, such as actinides, uranium, ^{90}Sr, ^{210}Po, ^{226}Ra, ^{3}H, ^{14}C and natural and anthropogenic gamma emitters. continuously subjected to internal verification through intercalibration tests.

Keywords

Radiation Protection, Radiochemistry, Intercomparison tests, Integrated laboratory

Introduction

All ENEA radiation protection activities are consolidated within the Institute for Radiation Protection (IRP) and are cross-functional, including with respect to existing or emerging strategic objectives. IRP collaborates with the Directorates of various Research Centers with a view to strategically positioning ENEA's activities, including at a regional level. The constant commitment to research allows ENEA to maintain its radiation protection and dosimetry services, provided both within and outside of ENEA, adequate and suitable to address current needs and emerging issues. The state of radiation protection at ENEA corresponds to the European state of the art, as it is constantly in touch with and engaged with international standards, also through the participation of its researchers and experts in scientific committees (e.g., ICRP, ICRU, IAEA, EURADOS) and standardization bodies (e.g., ISO, IEC, UNI). In 2010, during the reorganization of ENEA in its new role as an Agency, the Institute of Radiation Protection was identified as a first-level Unit and was given an internal organization to improve its efficiency and effectiveness in achieving its objectives. The Institute's structure is divided in four laboratories, including the Integrated Laboratory for Monitoring and Measurement of Radioactivity (IRP-MIR). This laboratory is located at the Casaccia, Saluggia, and Trisaia centers and brings together the Institute's main expertise and instrumental resources for radioactivity measurements on various types of samples. Using these resources, it ensures the

monitoring of internal contamination for personnel exposed to radiation at the ENEA centers and, also using resources available at other laboratories where necessary, ensures the provision of advanced technical services for radioactivity measurements and internal contamination. For the purposes of technical and scientific updating and the qualification of the measurements, research and development activities are primarily aimed at optimizing techniques for analyzing and measuring radioactivity in various matrices and individual monitoring procedures for internal contamination. The research and development activities were primarily focused on the study and testing of analytical and radiometric techniques for determining the radionuclide content in samples of various origins, primarily biological (excreta) of workers within individual monitoring programs, but also artificial and natural radionuclides in environmental samples (water, sediments). A further line of activity was aimed at designing rapid systems for measuring internal contamination to be used in the event of large-scale incidents involving the population.

Experimental

Internal contamination monitoring and measurement

Our integrated laboratory can provide internal contamination monitoring both for ENEA internal and external users, public and private where the services are subjected to a fee, owing to the capability to combine chemical and physical skills. The type of measurements, requiring a preliminary analytical treatment, are mainly: urine and fecal alpha spectrometry for actinides and ^{210}Po, urine beta counting for ^{90}Sr, ^{3}H, ^{14}C and gross beta, urine mass spectrometry for uranium isotopes and nasal mucus alpha/beta counting.

Urine and fecal alpha spectrometry

To assess possible internal contamination of ^{238}Pu, $^{239+240}$Pu and ^{241}Am in workers engaged in the decommissioning activity a proper monitoring program must include excretion analysis, since actinides do not have a sufficiently intense γ emission to be determined with *in-vivo* measurements. Although urine analysis alone is generally recommended, as individual variability in the representative 24-hour excretion sample is negligible, actinides are excreted primarily via the fecal route in the early stages of contamination, consequently, it seems appropriate to collect both types of excretion to obtain more accurate information on radionuclide intake [1, 2]. Our method allows the separation and purification of both plutonium and americium in order to determine their quantity in excreta starting from the same sample. This choice reduces errors due to the separation of the sample into two aliquots and the calculation of the intake starting from the measured activity is more accurate. Urine and fecal samples, marked with internal standards of ^{242}Pu and ^{243}Am to evaluate the chemical yield of the procedure, were conveniently mineralized and solubilized for separation by anion exchange chromatography with specific resins (commercially available) before being electroplated and measured by a high-resolution alpha spectrometry chain, for 600000 s. The combined use of the two cascade chromatographic methods allowed us to isolate the radionuclides of interest with a satisfactory decontamination factor and a constant chemical yield high enough to calculate a Minimum Detectable Dose (MDA) in blanks with good sensitivity. Besides, it eliminates the simultaneous electroplating of the natural isotopes of uranium and thorium in the plutonium fraction, without the need for a further step with another chromatographic technique and with a concomitant improvement in peak resolution [3]. Recently, fast determination methods for Pu and Am isotopes in small urine samples (50 mL) were developed [4] to give reliable dose evaluation in emergency situation, simplifying these procedures applied in the routinary monitoring.

In our laboratory we also use a technique for the determination of ^{210}Po in urine samples, based on mineralization and spontaneous deposition of polonium onto a silver disk. After the mineralization of the sample by covering the beaker with a watch glass and heating at a temperature no more than 90 °C to avoid the radionuclide evaporation, it is preconcentrated at the same temperature limit and diluted in hydrochloric media for the deposition in a heated water bath. The degree of the deposition is evaluated by adding ^{209}Po as a tracer. After waiting 24 h, the disk is measured by the same alpha spectrometry chain.

Urine beta counting

In case of contamination of β-emitter radionuclides it is possible to do a preliminary evaluation with gross β counting measurement or investigate a particular radionuclide, if the chemical species of the intake is known. Our laboratory is equipped to measure β counting with a proportional counter or liquid scintillation device. In case of urine as a matrix, pretreatment may be necessary to have the radionuclide in an environment suitable for the measurement. Every sample analyzed has generally a volume of 500 mL. For gross β counting urine samples are mineralized analogously as for actinide determination, but with particular attention to solubilize and rinse them with 4M NH_4OH to eliminate interfering alkali metals in the supernatant after centrifugation, like ^{40}K naturally present in human metabolism [5]. The precipitate is measured in our proportional counter, which allows us to measure ten samples simultaneously in different detectors. Self-absorption curves for each detector have been derived by different quantities of KCl weighted and deposited on aluminum planchets, to obtain different mass thickness standards. Experimental data (detection efficiency vs mass thickness) with mono-exponential functions has been fitted to obtain realistic efficiency, when samples do not have the same mass thickness of the standard.

Strontium in urine samples

For ^{90}Sr we have developed a method for its separation from other radionuclides, preliminary mineralizing the sample. For chemical yield the carrier used is stable strontium nitrate, while the extraction took place using a selective resin, consisting of 1.0 M 4,4'(5')-di-t-butylcyclohexane[18]crown-6 in 1-octanol. After passing the sample, appropriately mineralized, through a pre-packed column the radionuclide is eluted and separated from its daughter ^{90}Y, which can begin to grow again and becomes suitable for a Cherenkov effect measurement. By using a mathematical fit on experimental data following ^{90}Y ingrowth it is possible to determine its activity at the equilibrium with the mother ^{90}Sr, even before it has been reached, and so the activity of this last radionuclide. One-tenth of the eluate is collected to determine the chemical yield by AASS [6].
Lastly for ^{90}Sr we developed a rapid screening on a large number of sample, as required during emergency situations, to detect immediately and simultaneously many samples of people potentially contaminated. In this case from each collected sample, 30 mL aliquots are taken and transferred to centrifuge tubes, in which the radionuclide of interest is precipitated to separate it from the supernatant, which contains any other interfering radionuclides. From this dissolved precipitate, ^{90}Sr is separated and isolated with the same chromatographic method. After the collection of one-tenth of the eluate for the chemical yield, the sample is deposited and evaporated on a metal platelet to be measured by beta counting. This allows for rapid preliminary measurements of up to ten samples simultaneously. Considering that a limited number of counts due to ^{90}Y, which unavoidably has grown during the elapsed time from the ^{90}Sr separation and the end of the measurement (together

with the scarce counts of ^{90}Y that might not be retained by the column, already present in the collected sample partially for the excretion of metabolized yttrium and partially as a product of strontium decay), cannot be distinguished by ^{90}Sr pure counts, the evaluation of the activity can be considered as affected by an overestimation of no more than 10% [7].

Tritium in urine samples

For the determination of tritium in urine samples, it is necessary to previously discolor them with activated carbon to eliminate the color quenching. In fact, the measurement, starting from a quantity of 7 mL of urine, is performed by using an Ultra-Low Level Liquid Scintillation Spectrometer Quantulus 1220 (Perkin Elmer) and quenching effect must be avoided. Subsequently each sample is diluted with 3 mL of ultrapure water, added with Ultima Gold LLT and measured for 120 min. Measurement is repeated 3 times (in order to take into account the casual errors of counting) and arithmetic mean and standard deviation of net counts are calculated. The counting efficiency is obtained as the ratio between the mean net count of a reference source and its activity per unit volume. The activity of each sample is calculated as the ratio between the mean net count, subtracting the contribution of a blank sample, and the efficiency, divided by the volume of the sample. Since tritium is chemically similar to hydrogen, it can enter into human metabolism and be incorporated into organic matter. The dose from this organically bound tritium is more relevant, because it is not as easily eliminated as tritiated water. Therefore, it is mandatory to distinguish the contribution of the two different types of tritium. In our laboratory we determinate tritiated thymidine, as a marker of organically bound tritium in urine samples, always by liquid scintillation counting measurement. The activity of tritiated thymidine is calculated as the difference between the total activity and the tritiated water content, extracted by distillation of a small aliquot of urine (10-20 mL) in a glass Petri, covered with lid and heated on a hot plate. Droplets on the lid are collected with a transfer pipette until collecting a volume of 4-5 mL into a vial for addition of the same cocktail described above. Also in this case proper standard and blanks samples are used to calculate activity.

Carbon 14 in urine samples

The determination of ^{14}C in urine is a fundamental biological monitoring technique (bioassay) for workers at risk of internal exposure to this radionuclide (e.g. in biochemical or pharmaceutical research laboratories or nuclear plants). Carbon-14 is dangerous if ingested or inhaled. The main technique for determining ^{14}C activity in urine is Liquid Scintillation Counting (LSC) also in this case. Since urine is typically treated to reduce quenching (quenching of luminescence), a calibration curve, appropriately constructed with a series of quenched standards, accounts for the decrease in counting efficiency related to quenching due to urine color (see Fig.1). This curve shows the variation in efficiency as the quenching parameter SQPE varies; in liquid scintillation counting (LSC), SQPE stands for Spectral Quenching Parameter of the External Standard [8].

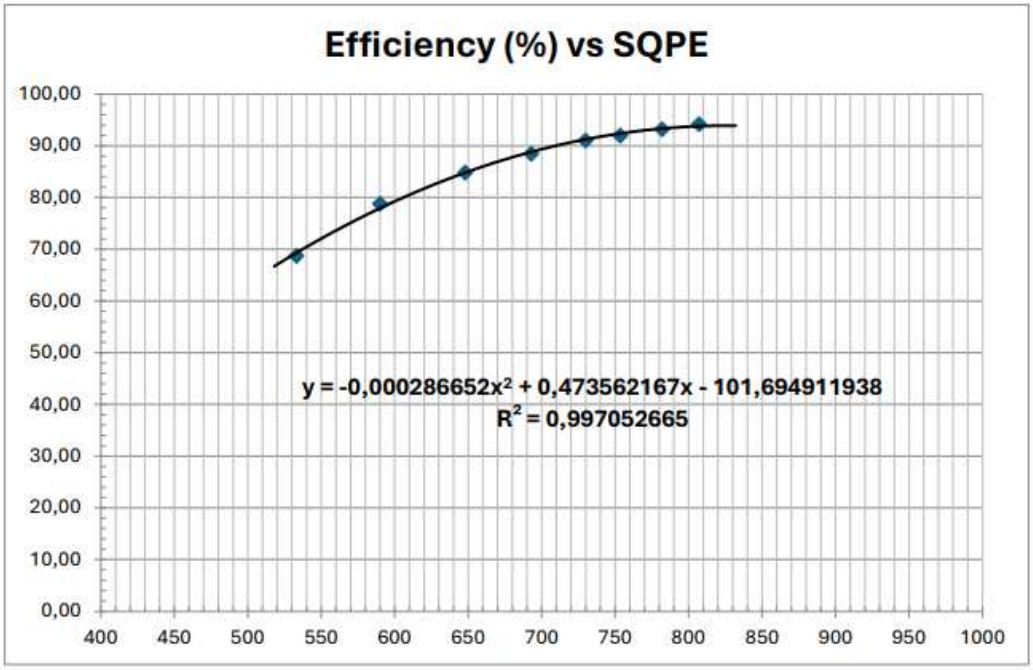


Fig. 1. Our procedure used for the determination of ^{14}C in urine involves adding 1 mL of urine diluted with water (1:4) to 10 mL of Ultima Gold LLT scintillating liquid. Each sample is measured for at least 30 min or even for 120 min for low-radioactive samples, repeating this measurement 3 times, as for tritium.

Determination of ^{234}U/ ^{235}U/^{238}U and ^{232}Th by ICP - MS

The ENEA IRP-MIR laboratory of Casaccia is equipped with an Agilent 7700 ICP–MS (Inductively Coupled Plasma Mass Spectrometer) powered by Helium Collision Cell and ASX500 autosampler to perform uranium and thorium isotopes determination as well as isotopic ratio $^{235}U/^{238}U$ and $^{234}U/^{238}U$ determination, for radiation protection purpose. For both biological and environmental samples, the matrix must be dissolved in diluted acid to be introduced by a Micromist concentric Nebulizer in a cooled spray chamber, creating a fine aerosol that is transferred to the Argon plasma generated through inductive coupling of free electros with rapidly oscillating magnetic field. The high-temperature plasma atomises and ionises the sample, generating ions which are then extracted through the interface region and into a set of electrostatic lenses called the ion optics. The ion optics focuses and guides the ion beam into the quadrupole mass analyser. The mass analyser separates ions according to their mass-charge ratio (m/z) and these ions are measured at the detector. The signal acquired by the detector is quantified by external standardization through a calibration curve for each isotope, with a very low instrumental Detection Limit (0,1 ppt for ^{238}U). The calibration range depends on the expected measured isotope amount: for ^{238}U in urine samples, it is usually between 1 and 20 ppt whereas in environmental samples it could be higher (up to 1 ppb or more) depending on the sample matrix and the dilution factor. Another advantage of the ICP-MS determination is the potentially simple sample pre-treatment, as in the case of uranium or thorium isotopes determination in urine samples. A simple dilution of the sample is sufficient to obtain reliable quantification, with a Method Detection Limit (MDL) about 1,2 ppt for ^{238}U.

Results and discussion

Qualification of the services provided is certainly one of the cornerstones underlying he Laboratory's activities. In addition to extensive internal work to standardize analysis procedures and verify measurement parameters, regular participation in intercalibration and intercomparison initiatives has been a regularly adopted and pursued practice for years. This is particularly important for types of measurements, such as radioactivity in biological and environmental samples. In the context of internal contamination monitoring for in vitro measurements, the primary tool for assessing the reliability of the performance provided by IRP Laboratories is regular participation in intercomparisons organized by the PROCORAD® Association (Association for the Promotion of the Control of Quality of Medical Biology Analyses and Radiotoxicology), probably the most recognized initiative of its kind at the international level.

Overall, as it has been the case for several years, the results obtained by IRP Laboratories in PROCORAD intercomparisons are considered widely satisfactory. This is further evidenced by the honorable mentions (TOP Lab) sent by PROCORAD for: tritium in urine (Casaccia and Saluggia laboratories, 2022 and 2025, Trisaia laboratory, 2023), uranium in urine (Casaccia laboratory, 2022 and 2024), plutonium in feces (Casaccia laboratory, 2022 and 2025), polonium in urine (Saluggia laboratory, 2023), actinides in urine (Casaccia laboratory, 2024, Saluggia laboratory 2025), strontium in urine (Trisaia laboratory, 2024).

The following figures (from Fig. 2 to Fig. 8) show the intercomparison results for biological sample analyses in the last four years for the three sites of our laboratory. We considered the absolute value of the uncertainty to evaluate the degree of deviation from the target value.

Generally, bias in determination of actinides in urine are lower than 10% both for the laboratories in Saluggia and Casaccia, with an episode of high bias for ^{241}Am and ^{244}Cm in 2022, due to resolution problems in the interpretation of the spectra. In 2023 bias higher than usual have analogously occurred in the determination of actinides in feces: in fact, generally bias have been around 10-15%. In this case too, reasons can be due to peak broadening which made difficult to distinguish between tracer and investigated radionuclide spectra.

Bias for ^{210}Po determination in urine has shown a relevant difference between 2023 and the two following years, with an excellent bias the first time (0-2%) for the two laboratories and bias even around 10% after; a possible explanation may be due to problems in the accuracy of the ^{209}Po tracer value.

Within determination of ^{90}Sr in urine the best results have been achieved by Trisaia laboratory where bias is not higher than 8% with a mean value of 6%., whereas the two other laboratories have bias generally over 10% (but less than 20%) with an average of 12%, probably due to different methods for evaluating chemical yield. Trisaia laboratory uses the common method of atomic absorption, while Casaccia and Saluggia uses ICP-mass spectrometry. Besides it is remarkable that Saluggia laboratory has alternated this last technique with the employment of the external standard, because the mass spectrometer is not present in the laboratory, but only in Casaccia where aliquots of the samples need to be sent and this cannot always be possible.

Determination of tritium in urine has generally proved low bias, except for that one in the organically bound tritium (OBT), for which the use of distillation before measurement in liquid scintillation is required in order to separate the free radionuclide in the water content from the one incorporated in metabolic chain. Considering that this value is calculated by the difference from total tritium activity and the portion obtained by distillation, the higher value of bias can be due to propagation of uncertainty.

Bias of ^{14}C are usually lower with an average of 3-4%.

Biases for ^{234}U, (234, 235, 238) for both mass and activity determination are generally less than 10%, often less than 5% in 2022, 2023, 2024 PROCORAD Intercomparison. The high BIAS values in the ^{234}U activity determination and ^{235}U mass determination could be justified by a likely inhomogeneity in sample distribution, these data are in any case related to Z-score less than 1, reported by PROCORAD association itself.

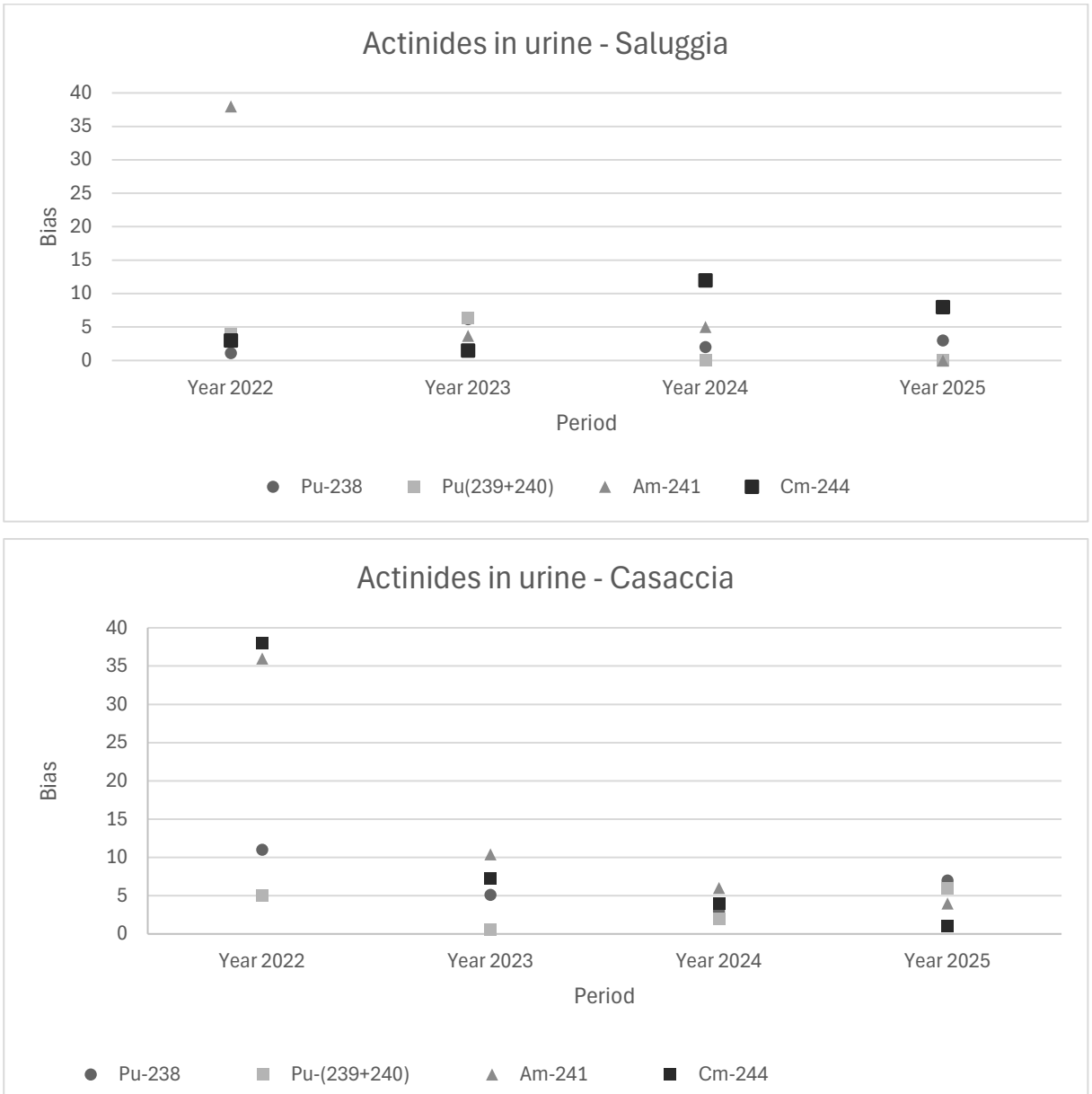


Fig. 2: Biases in PROCORAD intercomparison exercises for actinides determination in urine samples during period 2022-2025.

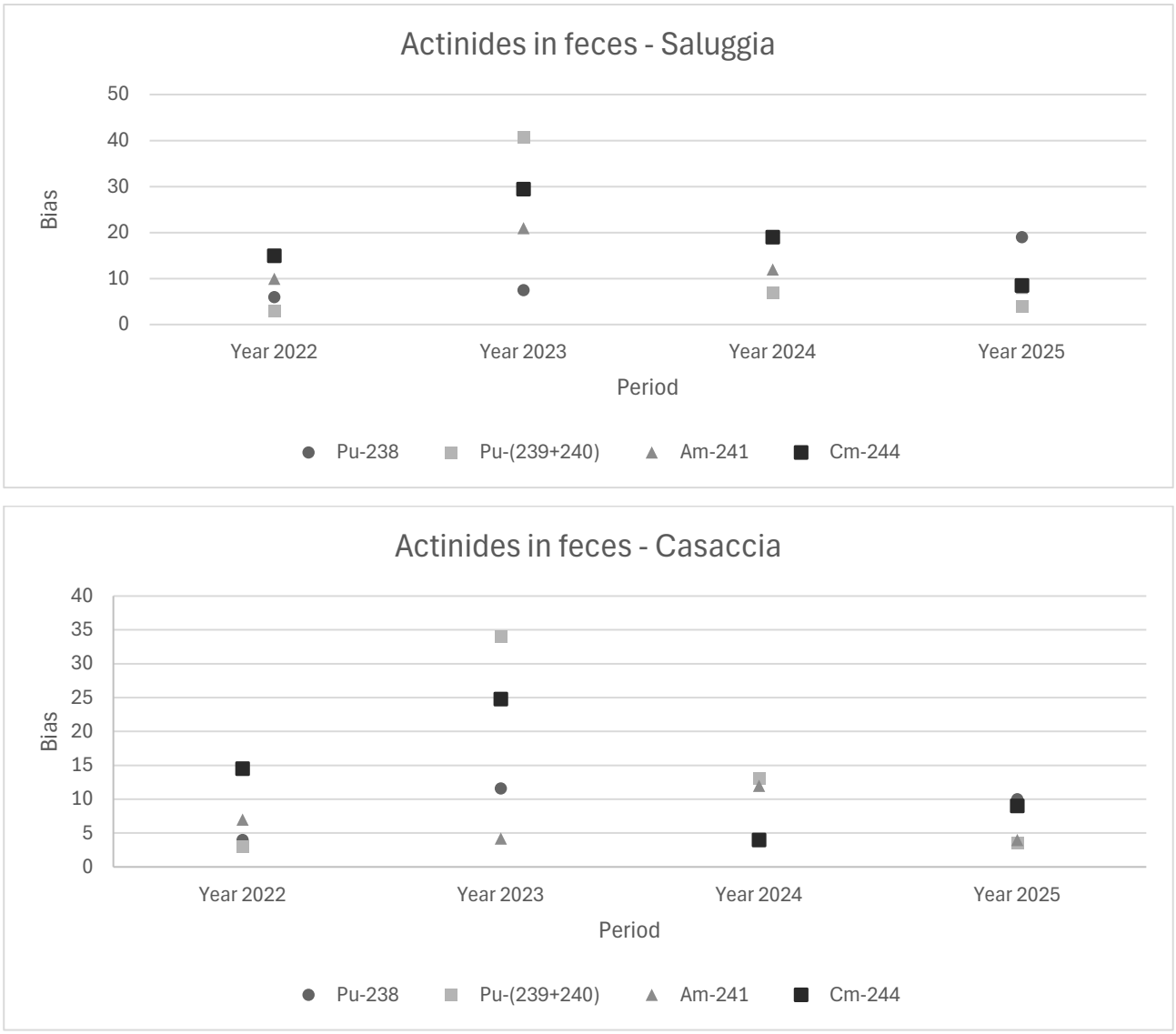


Fig. 3: Biases in PROCORAD intercomparison exercises for actinides determination in fecal samples during period 2022-2025.

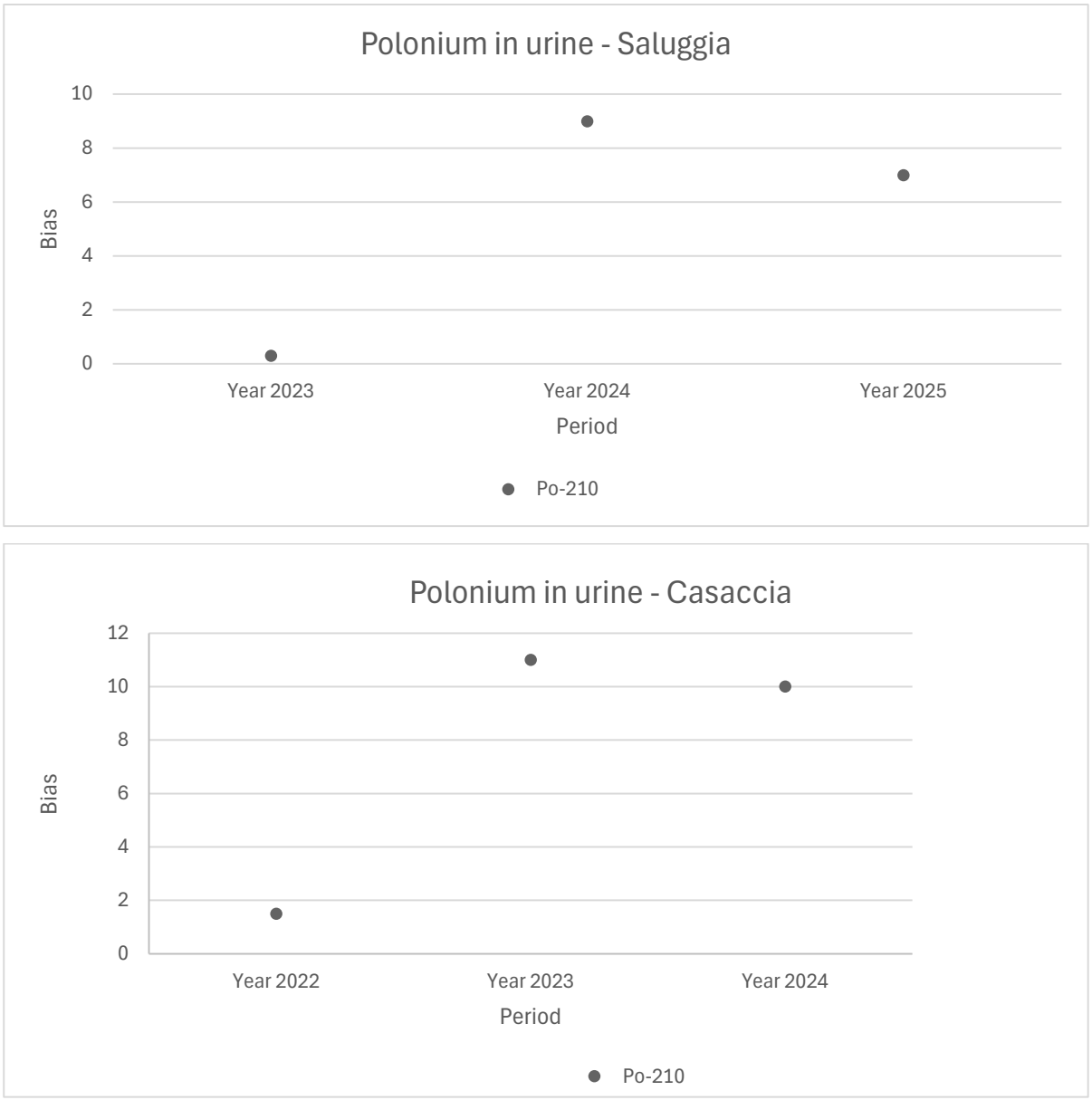


Fig. 4: Biases in PROCORAD intercomparison exercises for ^{210}Po determination in urine samples during period 2022-2025.

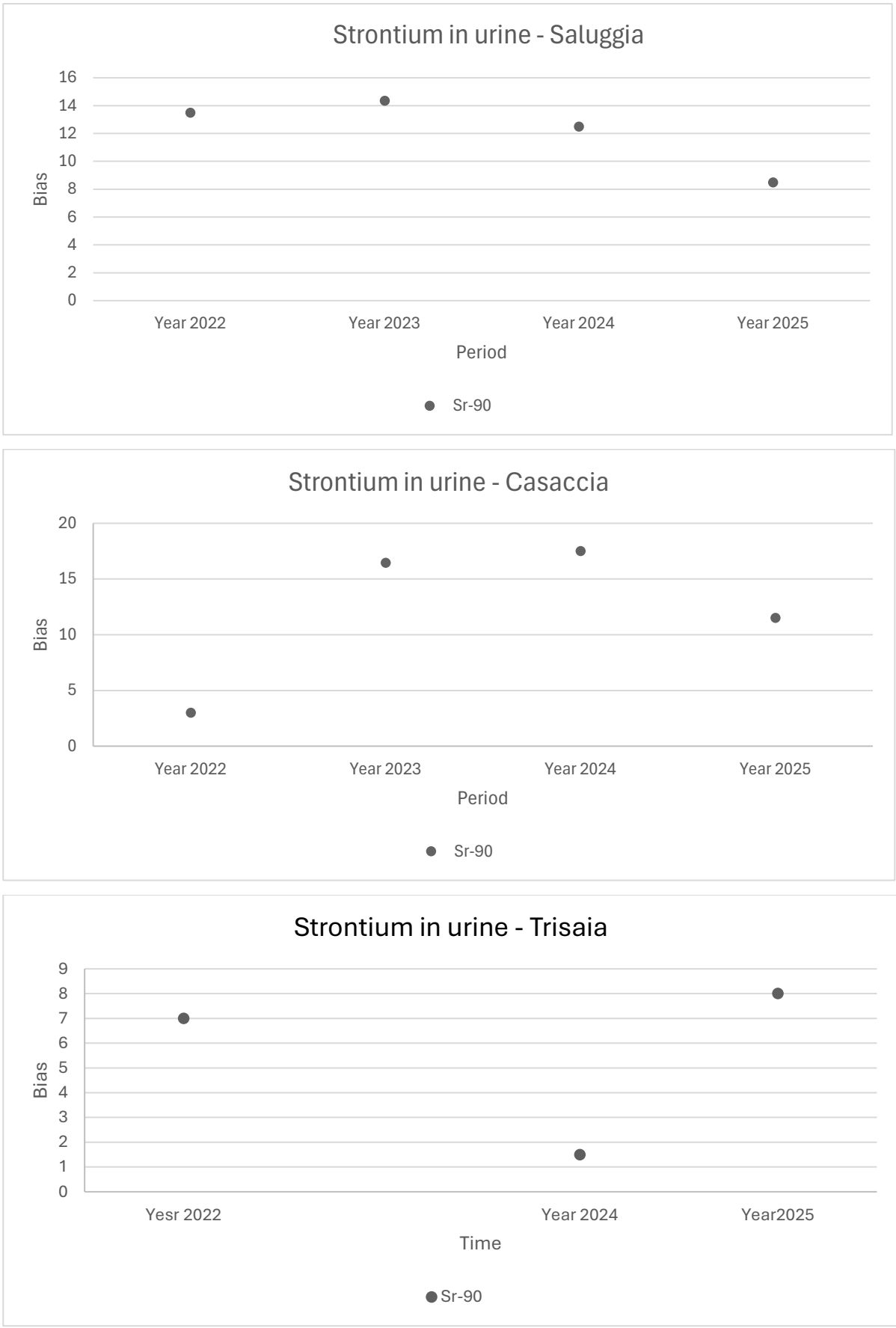
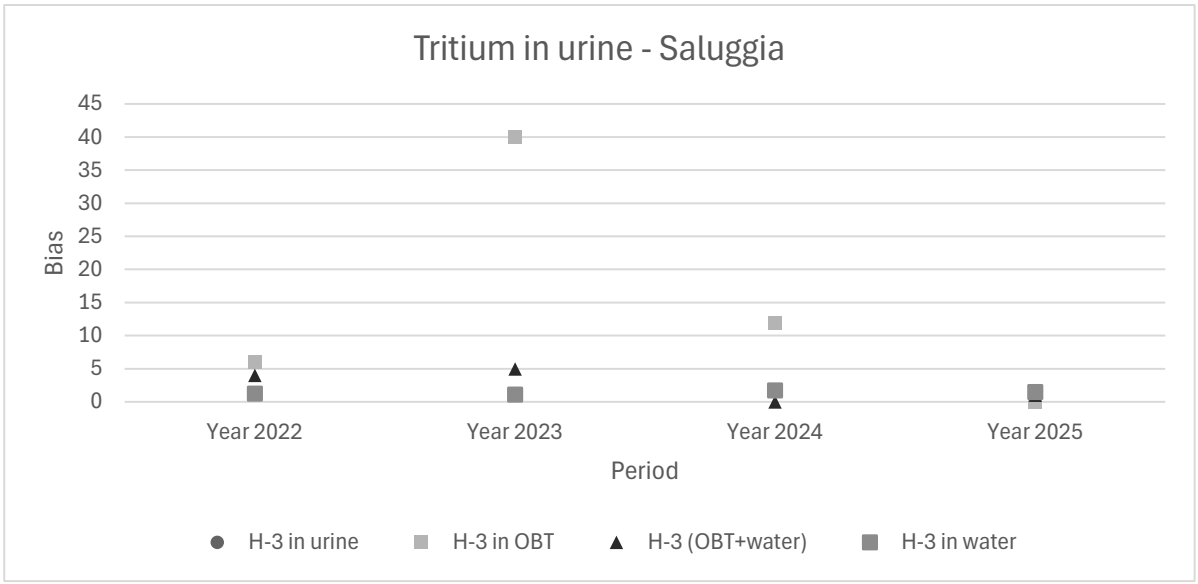
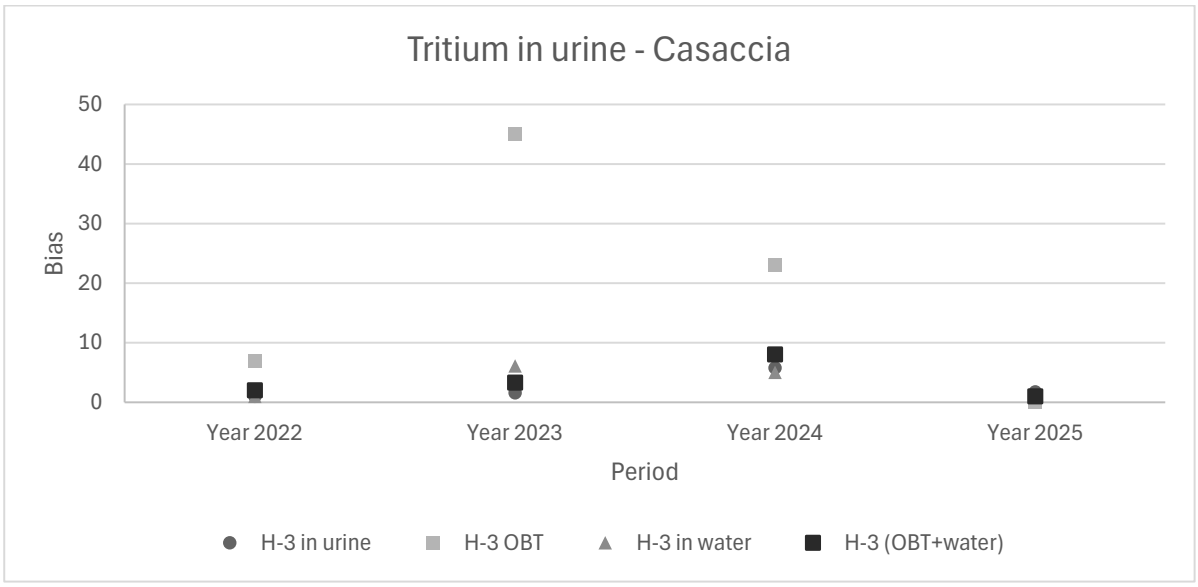


Fig. 5: Biases in PROCORAD intercomparison exercises for ^{90}Sr determination in urine samples during period 2022-2025.





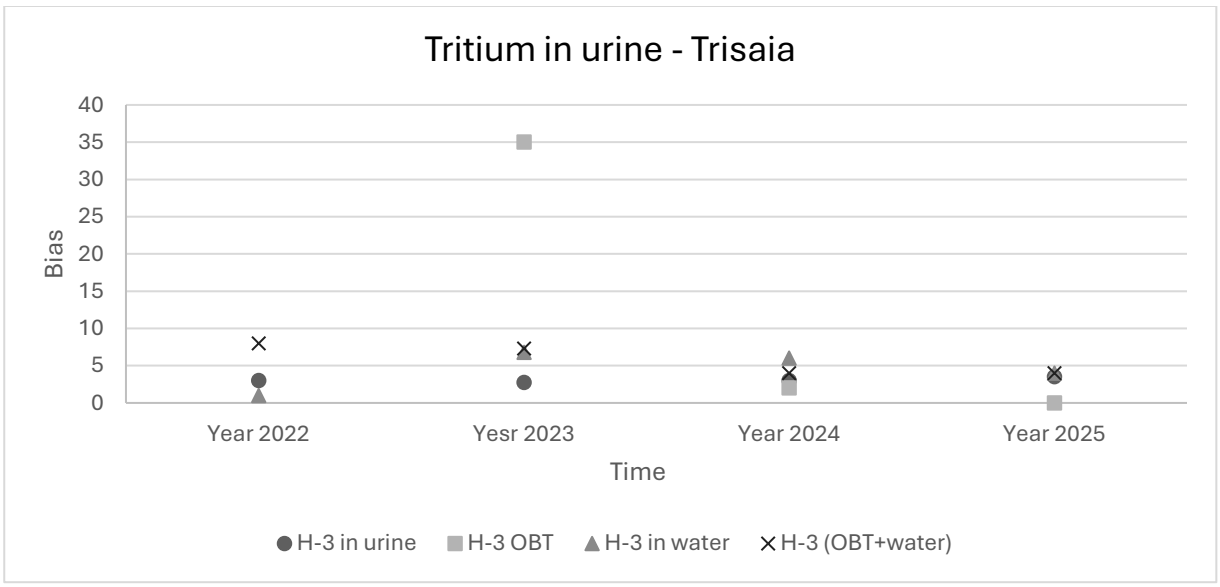
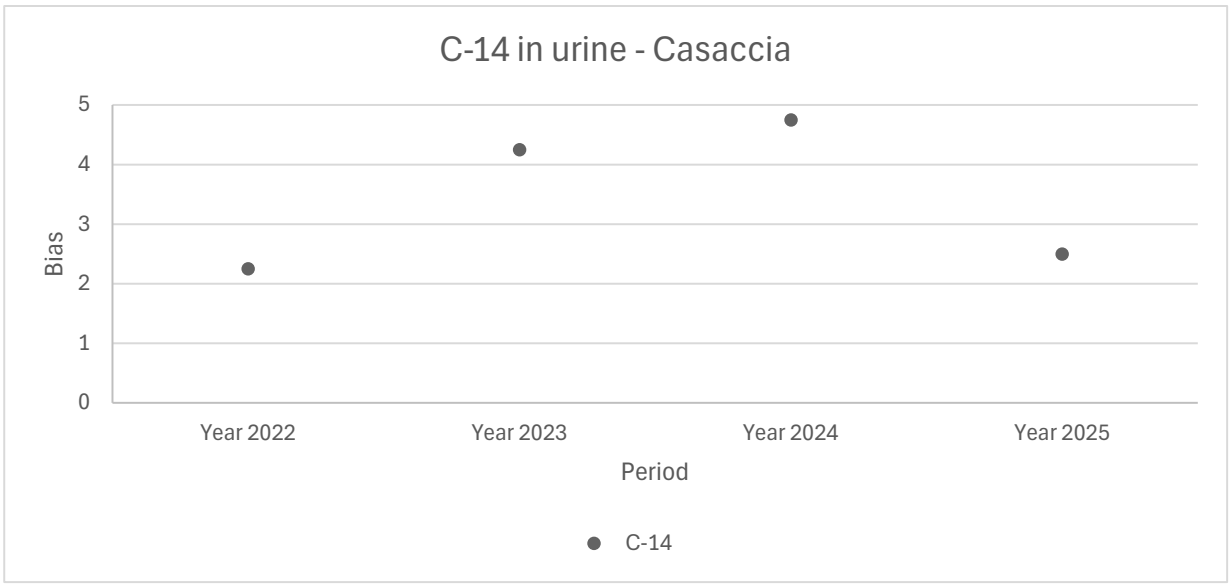


Fig. 6: Biases in PROCORAD intercomparison exercises for tritium determination in urine samples during period 2022-2025.



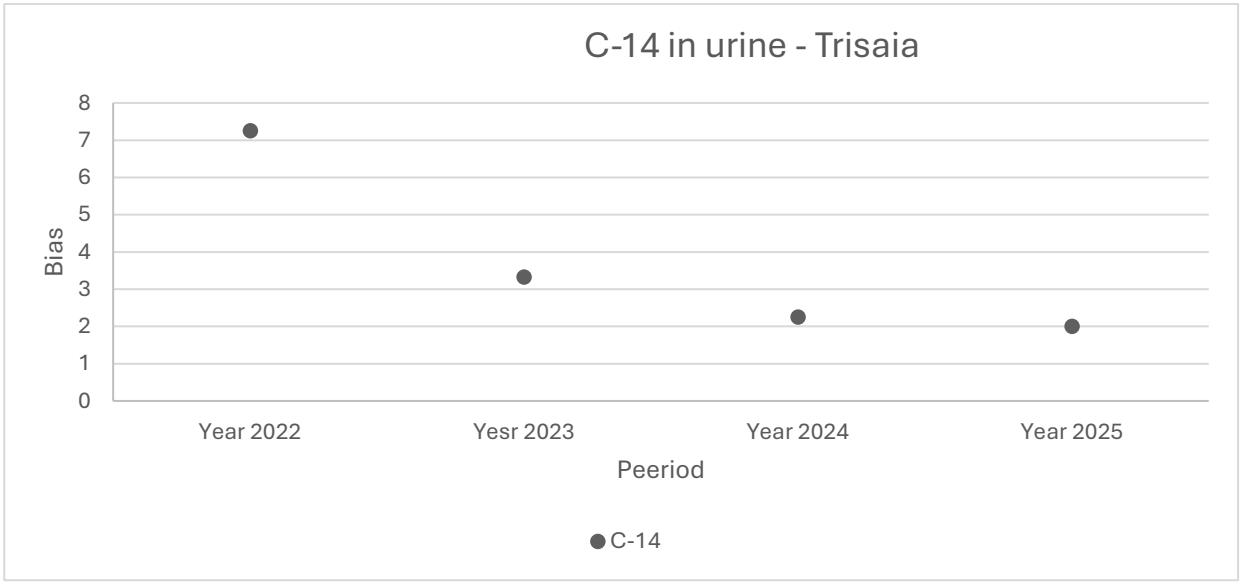


Fig. 7: Biases in PROCORAD intercomparison exercises for ^{14}C determination in urine samples during period 2022-2025.

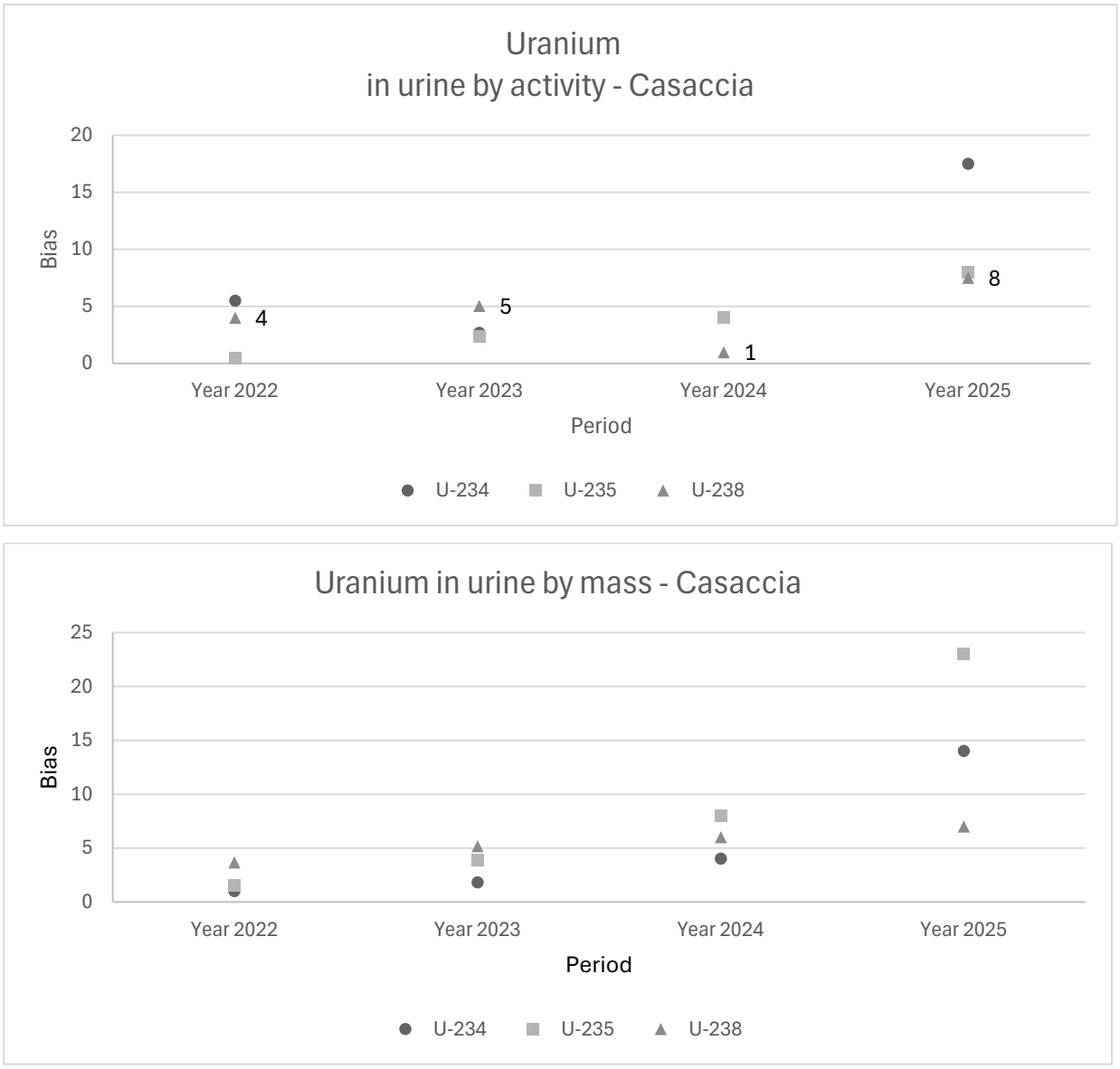


Fig. 8: Biases in PROCORAD intercomparison exercises for uranium isotope determination in urine samples during period 2022-2025.

Conclusion

The satisfactory results achieved in proficiency tests over the years proved the IRP-MIR Laboratories' competence, accuracy and reliability in different radionuclide analyses. With a view of continuous improvement, these exercises helped us to identify areas for improvement in routine analysis, such as optimizing sample preparation or treatment leading us to maintain high standards improving accuracy in measurements or reducing their uncertainties.
This made our laboratory a unique facility in Italy capable of satisfying every need for measurement and control of radioactivity for the safety of the population.

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