ENVIRONMENTAL RADIOACTIVITY MONITORING IN SPAIN.
WORKING TOWARDS A HIGHER QUALITY

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Abstract. The radiological environmental monitoring system in Spain has experienced continuous improvements aimed to the implementation of the last technology and to an adequacy to the recommendations of the European Union (EURATOM Treaty article 36). The management of the national environmental monitoring network is responsibility of the CSN (Nuclear Safety Council), and the performance of programs in these networks is carried out with the collaboration of laboratories in autonomic communities that provide the radioanalytical results in compliance with general criteria established by the CSN. Following the implementation of the national network, joined forces are being dedicated to improve the quality of the analytical results produced by laboratories and to achieve their international traceability to internationally recognised standards. CSN has required the development of quality systems in those laboratories and also, to maintain and verify the reliability of their results, organises in collaboration with CIEMAT periodical inter-laboratory test comparisons. Furthermore to ensure the quality and international comparability of the radiological evaluations, different activities are being developed; since 1998, the organisation of the biannual Workshops on the Quality in the Environmental Radioactivity Control in which relevant topics are presented and discussed. As a consequence of these Workshops and the annual feedback-meetings organised by CSN among network laboratories, three Working Groups have been established at a national level to harmonise criteria and methodologies used by laboratories. All these activities are performed in collaboration with relevant International Organisations. The paper describes the activities performed so far, and discusses the future trends to continue improving the quality of the environmental radioactivity monitoring in Spain.

1. Introduction

The monitoring of the environmental radioactivity in the European Union (EU) is carried out by Radioactivity Environmental Monitoring (REM) programmes, which have been established in the EU Member States to comply with legal national precepts established in accordance with the requirements of the EURATOM Treaty [1]. These programmes provide relevant information on radioactivity levels in all compartments of the biosphere to ensure that concentrations of radioactive materials do not constitute a risk to humans or the environment. The monitoring system consists of two complementary networks: the REM-dense network with numerous sampling points covering the entire Member States’ territory and the REM-sparse network with a limited number of selected locations.

The REM-sparse network was implemented by the European Union Member States to obtain data on actual levels of radioactivity, when the background levels of weapons’ fallout were decreasing and most of the data were reported as “less-than” values. Thus the sparse network requires that laboratories provide data with high sensitivity measurements and the highest achievable accuracy, to allow comparison of data sets for extended time periods and to follow the trends in the evolution of radioactivity levels.

The REM-dense network was fully established in Spain by the CSN in 1992, and according to EU recommendations the sparse network was implemented in the year 2000; this is a subgroup of the former one, where for each region of the national territory (4 in the peninsula and 1 in the Canary Islands) and media, exists one representative sampling location and it is integrated by 5 laboratories (see FIG.1). The management of these national environmental monitoring networks is responsibility of the CSN, and the performance of programmes in these networks is carried out with the collaboration
of laboratories in the autonomic communities that provide the radioanalytical results in compliance with general criteria established by the CSN.

Since the first monitoring network was established, the CSN has always been enforcing laboratories to improve the quality of their results, following the implementation of the national network special attention is being paid to enhance the detection capability of the measurement systems of the laboratories, and to increase the accuracy of their results to achieve traceability to international standards. Several actions are being undertaken to reach these objectives, a short description of the activities performed in collaboration with relevant institutions is given below.

2. On-going Activities

The quality of the information coming from these REM programmes as basic data for the assessment of the potential risk to human or the environment, has been a continuous CSN concern and to evaluate regularly the reliability of the data produced by laboratories, the CSN organises in collaboration with CIEMAT periodical inter-laboratory test comparisons. For the last six exercises a methodology for the assessment of the laboratory’s performance has been implemented based on the recommendations of the ISO/IUPAC/AOAC international protocol [2] and the ISO-43 guide [3]. This protocol recommends the use of a reference material as a test sample (which assures traceability to international standards) and the z-score statistical treatment to assess their performance; this procedure provides laboratories with a transparent and objective means of assessing and demonstrating the reliability of their results, and allows to set the performance criterion at the routine working level of laboratory participants.
The established system has shown over the years to be an adequate tool to improve the quality of the results, enabling laboratories to evaluate the accuracy of a particular analytical method and make appropriate modifications to their routine procedures. FIG.2 resumes the exercises performed so far, samples utilized are similar in composition and activity levels to the ones routinely analyzed in the REM programs. The number of participants has been increasing as some Laboratories of REM networks have joined these exercises as an external quality control of their performance. These are mainly research laboratories in which a quality system is not so well established, and the reliability of their results is also important [4] for environmental impact assessment, as they provide relevant parameters on the processes governing the transfer of radionuclides in the biosphere for the predictive models.

FIG.2. Interlaboratory exercises performed among REM laboratories.

As a consequence of the annual feedback meetings organized by CSN with the participation of REM and these other laboratories, it came into view a lack of homogeneity in criteria and formulation applied by different laboratories. Some global activities have been established in order to promote the required harmonization at a national level according to international standards, i.e.: to organise workshops with international expert opinions, to create working groups to adapt methodologies to international criteria and agreed upon fundamental principles evaluation of uncertainties, procedures, the use of adequate radioactive standards (traceability), and detection limits.

2.1. Quality System in Laboratories

If environmental regulators are to make appropriate decisions these have to be based on reliable facts, including environmental measurements that regulators, industry and public can have confidence in. As stated above the quality of the results obtained in the environmental monitoring programmes has been a constant concern of the CSN which has required network laboratories to implement strategies to
improve the quality of the results they produce. To meet these requirements laboratories must establish quality assurance systems including relevant requests in standards or regulations such as the use of validated methods of analysis, internal quality control procedures, participation in external proficiency tests schemes, accreditation (ISO 17025) [3], and establishing traceability of the measurements to a given standard.

The CSN has developed over the time a general policy to assist the laboratories in achieving this goal, starting in 1987 with the development of a document on the quality assurance to be implemented in the monitoring programmes, which was discussed with 12 laboratories taking part at the time in the surveillance network. In 1992 when the CSN monitoring network was fully established, some main elements of the quality system were incorporated. In the collaboration agreements the laboratories were required to develop, prior to incorporation in the network, a technical document including a description of sampling, detection and measurements equipment; the sampling, analysis and measurement procedures used by the laboratory; a quality assurance programme; and the participation in the analytical inter-laboratory exercises organised by the CSN.

In 1997 the CSN requested the participant in the REM networks to formally implement a quality system and develop Quality Manuals and a programme for establishing, implementing and optimising them, since the application of a quality system that completely integrates the organisation’s structure, responsibilities, procedures, processes and resources required for suitably managing quality is an effective way to achieve the required goals.

In order to verify that the established quality programmes are properly enforced, internal controls are introduced into the organisations and external actions are taken, such as the above mentioned inter-laboratory exercises and audits.

2.2. Organisation of Workshops on the Quality in the Environmental Radioactivity Control

The lack of specific information regarding accreditation for radioactivity laboratories led to organise a forum where scientists involved in radioactive measurements could gather experts from different activities regarding the necessary steps for accreditation and seek advice on the requirements to achieve an adequate quality system in their laboratories. The first meeting was organised in Bilbao by the SNE (Sociedad Nuclear Española) in 1998 [5], the come out of the workshop presented many other areas, besides accreditation, in which a deeper study and harmonization was required (sampling procedures, radiochemical methods, the use of adequate certified standards for traceability, methodologies and criteria to evaluate measurement uncertainty, limits of detection/decision, different terminology and concepts,...). It was concluded that specific working groups should be established at a national level to endeavour harmonisation of different criteria.

Two years later, in year 2000, a second meeting was organized under the auspices of the SNE, aiming to follow up the activities initiated in Bilbao, and to continue with actions initiated to achieve the intended harmonization. The workshop was held in Salamanca and the main topics discussed were “quality assurance in radioactivity determinations”, international activities regarding this matter and the working groups formed two years before presented the results obtained after a first period of operation.

The increased participation of scientists and the growing interest in quality assurance processes, led to the decision of establishing the bi-annual organization of the workshops, and subsequently in 2003 a third meeting took place in Valencia, this time as joint effort of the Spanish Nuclear Society and the Spanish Society for Radiological Protection (SEPR). The meeting was mainly focused on gathering experiences from laboratories already accredited or in process, also REM-laboratories from other...
Member states were invited to participate and exchange experiences; in addition the role that accreditation is playing in other countries was described. The coordinators of the working groups presented the development of their tasks and described on-going activities and documents produced as a result of their work.

These meetings, where information exchanging among scientists is being produced, have been consolidated as a means to reach the required harmonisation of criteria and methodologies; and the following meeting will be held in Sevilla during 2005.

2.3. National Working Groups for standardization

The necessary comparability of the results among different Member States’ laboratories involves, further to the implementation of a quality assurance program, the harmonisation of criteria, sampling procedures, calculations, or the reporting of results, agreed upon fundamental principles and international standards. Shortly after the Bilbao 1998 Workshop, a REM-network laboratories meeting was organized at the CSN and three Working Groups were established,

− Standards for monitoring procedures
− Radioactive standards
− Evaluation of uncertainties

2.3.1. Standards for monitoring procedures

Measuring radioactivity in environmental samples involves a complex and lengthy process that includes several steps from collection and preparation of representative samples and their chemical analysis to calibration of measurement equipment. Representative samples must be taken and the treatment in the laboratory should guarantee that the final data reproduce as close as possible the contents of radioactivity in the environment. The application of standards in the whole process is one of the key issues to accomplish this goal.

In the Working Group established to develop standards for monitoring three subgroups were set up to develop procedures for the main stages of the environmental sample radioactivity measurements process: sampling, sample preservation, analytical methods and preparation and measurement equipment. On the basis of issues considered to be of higher priority, these group started working on development of soil, water, and air sampling procedures, analytical procedures for $^{90}$Sr in soil and sediments and beta activity in water, along with procedures for calibrating gamma, and alpha spectrometry equipment. It was considered convenient that these group carried out their activities in co-ordination with the AENOR, giving rise to several Spanish Norms, as stated below, where the procedures and norms already produced by these groups are listed.

Sampling for measuring environmental radioactivity.
- Soil. Superficial layer. (Published as UNE 73311-1)
- Particulate and radio iodine in air.
- Continental and sea sediments

Conservation and handling of samples for measuring environmental radioactivity.
- Soil sample (Published as UNE 73311-5)
- Particulate filters and charcoal samples

Analytical methods for determining environmental radioactivity
- Gross beta in water by proportional counting (Published as UNE 73311-4)
- Residual beta in water by proportional counting (Published as UNE 73340-2)
- Determination of $^{89-90}$Sr in soils and sediments
Measuring equipment

- Gamma spectrometry with semi-conductor detectors (Published as UNE 73350-1)
- Alpha spectrometry with semi-conductor detectors (Published as UNE 73350-2)
- Liquid scintillation detectors (Published as UNE 73350-3)

These groups have also analysed the applicability of the ISO standards on water quality to sampling conservation and handling of samples, concluding that they can be directly applied by laboratories, not requiring further development. Present activities include developing procedures for sampling $^3$H and $^{14}$C in air, rain and ground water, along with analytical methods for determining $^{89-90}$Sr in particulate filters, water and organic samples.

2.3.2. Radioactive Standards

Some discrepancies between values reported by participating laboratories and reference values used in intercomparison exercises are found to come from dispersion in standards usage and basic data, such as half-lives, branching ratios, etc, used to obtain activity values from recorded countings.

Thus it seemed of capital importance to harmonise data and standards usage and to this end a Working Group (GPR) was created to elaborate a harmonisation manual [6].

This, which will be published in the very near future, contains a scope of geometries and matrices used by laboratories in surveillance programs as well as of those radionuclides of interest for each program.

In a first instance only $\gamma$-spectrometric measurements have been considered and for this recommendations on radiochemical methods to prepare secondary standards and methods to assess a proper performance of detectors, checking for instance the performance of pile-up rejection systems, as well as to correct for physical effects, as coincidence summing for instance, influencing the result have been recommended. Finally a method to choose among different calibration cocktails has been developed.

This manual has been elaborated to meet the requirements of national and international standards in order to reach a general validity harmonisation.

2.3.3. Evaluation of uncertainties

Results from radiochemical determinations always involve uncertainty, every measured value should be accompanied by an statement of its uncertainty estimate, as it constitutes an implicit expression of the accuracy and precision associated to the result. The confidence to be placed in results is possible only if a quantitative and reliable expression of their relative quality, the associated uncertainty, is assessed.

REM programmes, involve large numbers of results from environmental radioactive determinations being compared to basic standards or to be within specific limits. These determinations are frequently performed at levels where the radionuclide of interest cannot be distinguished from natural background levels, and the relative uncertainty associated with the result tends to increase. Playing thus an important role for interpretation of analytical data and consequently in making decisions related to risk assessment.

This Working Group (GTINC) was created to achieve harmonization of criteria, formulation and the reporting of results based in international standards [6], [7], aiming to establish a uniform methodology for the evaluation of uncertainties in environmental determinations, to be applied by all
the network laboratories. The work performed has been reflected in a document [8], which was early distributed among network laboratories via Internet. The application of the methodology described in the document has contributed to enhance comparability of their results and to check their routine procedures making the appropriate modifications.

Due to the low activity levels found in the environment, measurement of radioactivity has to be made close to detection limits of the detection systems and as stated before the relative uncertainty tends to increase to the point where the (symmetric) uncertainty interval includes zero. This region is typically associated with the practical limit of detection for a given method, this is a “conflicting region” where some confusion exists due not only to the difficulty of establishing decision/detection levels, but also due to the numerous existing criteria, terminology and formulation.

The GTINC conscious of this problem started working in this area by reviewing all existing bibliography on the subject, and gathering opinion from international experts and at present is preparing a document where a common methodology and terminology be established agreed upon fundamental principles and international standards [10], [11], [12].

3. Conclusions / Future trends

The radiological protection of the environment and public can only be assured on the basis of reliable and traceable measurements to International standards. The reliability of the assessment obtained from these programmes requires that laboratories producing the analytical data be able to provide accurate results, traceable to internationally recognised standards, and for their quality to be adequately demonstrated and documented.

The actions developed up to now, aiming at the achievement of a higher quality in REM programmes have revealed the following progress in REM-laboratories:

- Increased analytical response capability
- Improvement of their analytical and measurement capacities: Calibration systems, lowering detection limits
- The assessment of the uncertainty associated to each radiochemical procedure utilized by laboratories has provided a mean to review and refine their methodologies
- The use of reference materials as a test sample in the interlaboratory exercises has contributed to improve the accuracy of the results and therefore their traceability

Also the settlement of national Standards on radioactivity measurement, will contribute to improve the performance of laboratories and the comparability of their results.

The experience from the described activities has shown that agreed methodologies developed on the basis of internationally accepted procedures, as those contained in international standards, constitute a valuable recourse to reach the aimed goals. The organization of periodical meetings among scientists from national REM-laboratories is an efficient way that contributes to achieve the harmonization of criteria and methodologies.

To enhance the reliability of the environmental impact assessment models, it should be promoted the collaboration of research laboratories into this bi-annual meetings to provide exchange of experiences between experimentalist and modellers.

The necessary comparability of the national laboratory results among different Member States’ laboratories implies more involvement in international committees for standardization.
Future trends in society regarding radioactive measurements would focus on the development of new methodologies to assist in the forthcoming decommissioning of Nuclear installations, where also new criteria should be applied.

In view of the inclusion of new EU Member States, a diffusion of the harmonised criteria and methodologies should be envisaged to maintain comparable measurement results in environmental radioactivity surveillance.

Acknowledgements

To network laboratories and the members of the working groups for their dedication and competence to achieve the intended goals.

4. References