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## Radioactivity monitoring in foodstuff and drinking water - An overview of performance of EU laboratories based on interlaboratory comparisons

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#### ABSTRACT

A compilation of the reported results of food and drinking water related interlaboratory comparisons conducted by the Joint Research Centre Institute for Reference Materials and Measurements is presented in this paper. An overview of analytical methods used by the participating laboratories, their overall performance as well as the evaluation of individual laboratories is assessed. The results show that the quality of input data into Radionuclide Environmental Monitoring database (REMdb) of the European Commission is acceptable but in some cases improvement is necessary. Harmonisation of the results obtained by different laboratories involved in REMdb and other European Union monitoring networks is worth the effort.

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#### 1. Introduction

Consumer protection and safe food production are among the main tasks of the legal authorities in every country. The food quality can be influenced by many factors and one of them is certainly the level of radioactivity, especially after nuclear emergency situations, such as the Fukushima-Daichi accident. Detailed knowledge of the activity concentrations of radionuclides in foodstuff is necessary to determine the effective dose to the population from the food chain, since the ingestion of food is the most significant route of radionuclide intake for the public (ICRP, 1992). Therefore it is essential to have emergency response procedures in place and well known to all participants in the process, from both the legal and the regulatory point of view. The laboratories involved in routine measurements of the radionuclides in each country of the European Union (EU) are an important part of these procedures, therefore, it should be guaranteed that the results they provide are of highest scientific value and that they can be trusted.

The EU, in its effort for reduction of radiation exposure to the minimum achievable level, has already taken steps leading to a

satisfactory level of emergency preparedness. The Euratom Treaty, signed in 1957, in the Article 35 states that Member States (MS) of the EU shall conduct a continuous monitoring of the environment and that European Commission (EC) has a right to control the performance of the facilities conducting the measurements (Euratom, 2012). Another legal act related to the monitoring of environment is the Commission Recommendation No. 473/2000 (Euratom, 2000). This document points out that monitoring of foodstuff should be carried out on a regular basis in all MS and networks designed for the purpose should be established (Máté, Sobiech-Matura, & Altzitzoglou, 2014). Also in the view of the recently adopted 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 it is of crucial importance to establish a network for monitoring the water intended for human consumption and to make sure that the quality of this water is in compliance with the limits set up by the directive (Euratom, 2013). All data concerning the radioactivity concentrations in various environmental matrices are collected and available for the general public via the Radioactivity Environmental Monitoring data base (REMdb) maintained at the Joint Research Centre Institute for Transuranium Elements (JRC-ITU).

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In order to evaluate the performance of laboratories involved in routine and emergency measurements of the environment and foodstuff and reporting their results to REMdb interlaboratory comparisons (ILCs) are organised biennially on request of EC Directorate General for Energy (DG Energy). These ILC exercises were originally organised by the French Office de Protection contre les Ravonnements Ionisants (OPRI, reorganised and renamed as Institut de Radioprotection et de Sûreté Nucléaire, IRSN) starting in 1991. Since 2003 the EC Joint Research Centre Institute for Reference Materials and Measurements (JRC-IRMM), as the provider of pro-active advice to the EC services on emergency preparedness, has undertaken the organisation of the ILCs (Wätjen, Spasova, & Altzitzoglou, 2008). Investigations carried out currently are focused on the harmonisation of the radioactivity measurement system in the EU. This study will allow spotlighting some problems with providing accurate measurement results encountered by laboratories monitoring radioactivity across the EU.

#### 2. Interlaboratory comparisons program

In the typical cycle of an ILC exercise for MS laboratories at first a selection on the matrix and the radionuclides is made, and agreement by the Euratom Treaty Art. 35–36 expert group consisting of representatives of all EU MSs is gained (Euratom, 2012). Then the same experts nominate monitoring laboratories in their respective countries to participate at the ILCs organised by the JRC-IRMM. These laboratories are routinely measuring radioactivity in the environment and foodstuff and they are reporting results to REMdb.

The matrix material is either prepared by the JRC-IRMM or purchased, as it may be a commercially available item (e.g. water with elevated natural content of <sup>234,238</sup>U or Certified Reference Material (CRM) produced by an external CRM provider outside the institute). The establishment of the reference value for the radionuclides of interest is conducted by JRC-IRMM, often with the participation of one or more national or international metrology institutes. JRC-IRMM contacts the ILC participating laboratories and samples are being sent to them. The last two phases of an ILC are evaluation of the results and reporting. After conclusion of the ILC the results are published. In all phases, the anonymity of the participating laboratories is guaranteed.

The overview of the results obtained by the participating laboratories of four ILCs is presented in this publication. In the first two ILC exercises organic matrices were used, and in the last two water intended for human consumption. On the basis of the previously conducted exercises it was attempted to identify ways to improve measurements carried out and methods used by the laboratories, by evaluating their performance.

#### 2.1. Organic matrix ILCs

Two ILCs using organic matrices were organised at the JRC-IRMM. The first one was the ILC on the 'Measurement of <sup>137</sup>Cs, <sup>40</sup>K, and <sup>90</sup>Sr in milk powder' ILC organised in 2005 (hereinafter referred to as Milk powder ILC). The material was milk powder CRM purchased from IAEA. The milk used for the production of this material originated from contaminated feed metabolised by cows in the Chernobyl area after the reactor accident in 1986. The reference values were established at JRC-IRMM. These reference measurements were conducted in the frame of the project leading to upgrading of Analytical Quality Control Services intercomparison materials to reference materials. The total number of participants from EU MS in this ILC was 61. This ILC is described in detail in Wätjen, Spasova, and Altzitzoglou (2008) and in Wätjen, Spasova, Altzitzoglou, Emteborg, and Pommé (2008).

The second organic matrix ILC exercise was the one conducted in 2011 on the '<sup>137</sup>Cs, <sup>40</sup>K, and <sup>90</sup>Sr measurements in dried wild bilberry powder' (hereinafter referred as Bilberry powder ILC). Bilberries were collected in northern Ukraine, in an area affected by the Chernobyl reactor accident. The material after preliminary drying and sorting was shipped to JRC-IRMM for further processing (Wätjen et al., 2015). It is now available for purchase as a certified reference material IRMM-426 Wild Berries; it is certified for the activity concentration of <sup>137</sup>Cs, <sup>40</sup>K, and <sup>90</sup>Sr. The reference values were established in the CCRI(II)-S8 supplementary comparison "wild berries" on the basis of results from nine National Metrology Institutes and international organisations. The number of participants of this ILC reached 78 laboratories from EU MS (Meresova & Wätjen, 2013; Wätjen et al., 2014).

#### 2.2. Water-related ILCs

The new European Council drinking water directive laying down the requirements for the protection of the health of the general public with regard to radioactive substances in water intended for human consumption (Euratom, 2013) includes derived levels for <sup>226,228</sup>Ra and <sup>234,238</sup>U. It also contains limits of detection for the gross alpha/beta, <sup>226,228</sup>Ra and <sup>234,238</sup>U measurement methods. In the frame of this directive two ILC exercises on radionuclide measurements in drinking water were organised by JRC-IRMM. The first one, conducted in 2008, was on the 'Measurement of <sup>226,228</sup>Ra and <sup>234,238</sup>U in mineral waters' (hereinafter referred as Mineral water ILC). The water used for that exercise was bottled mineral water purchased from three different companies. Three different samples were prepared, named W1, W2, and W3. In the sample W1 laboratories were asked to determine all four radionuclides, whereas in sample W2 only <sup>226,228</sup>Ra, and in sample W3 only <sup>234,238</sup>U. In total 47 laboratories participated in this ILC. The reference values for these samples were determined by a consensus between two independent expert laboratories, JRC-IRMM and Bundesamt für Strahlenschutz (BfS), Department for Radioprotection and the Environment, Berlin (Wätjen et al., 2010).

Since gross alpha-beta measurements are used for screening purposes in many fields, such as environmental monitoring and industrial applications, another water-related ILC was conducted in 2012 (hereinafter referred as Drinking water ILC) (Jobbágy, Wätjen, & Merešova, 2010). For this ILC exercise, the participants were asked to measure the gross alpha and beta activities in the distributed samples of water intended for human consumption (drinking water). Three samples were prepared, named WA, WB, and WC. Samples WA and WB were commercially available mineral waters, and sample WC was spiked water prepared at JRC-IRMM. The isotopes contributing to the alpha particle emission from sample WA consisted of natural isotopes of uranium (234,238U), and for sample WB <sup>226</sup>Ra. The naturally occurring isotope of potassium  $(^{40}\text{K})$  was determined by  $\gamma$ -ray spectrometry to be the main betaparticle emitter in samples WA and WB. Finally, sample WC was Type 2 water obtained from a Millipore ELIX-35 system acidified using nitric acid with added mixture of inorganic salts and spiked with <sup>90</sup>Sr/<sup>90</sup>Y and <sup>241</sup>Am. Samples WA and WB were not acidified in order to simulate the routine conditions in which sample is obtained by a monitoring laboratory from a manufacturer without any pre-treatment. Details regarding the samples selection and treatment can be found in Jobbágy, Dirican, and Wätjen (2013). The reference values were evaluated on the basis of measurements by three independent laboratories applying four routinely used methods (Jobbágy, Merešova, & Wätjen, 2014; Jobbágy et al., 2015). Overall 71 EU laboratories submitted results for this ILC, and a detailed description can be found in Jobbágy et al. (2014, 2015).

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