



Final Publishable Summary for 16ENV10 MetroRADON Metrology for radon monitoring

Overview

The aim of this project was to develop reliable techniques and methodologies to enable SI traceable radon activity concentration measurements and calibrations at low radon concentrations. The results of the project have been targeted at the implementation of the European Council Directive 2013/59/EURATOM (EU-BSS), one aim of which is to reduce the risk of lung cancer for European citizens due to high radon concentrations in indoor air. The calibration methods and measurement techniques developed in the project have started to assist EU member states in the establishment of their national radon action plan, which is required under the EU-BSS.

Need

Radon is a radioactive, colourless, odourless, and tasteless noble gas, which occurs naturally through geological-based processes. Despite its short half-life of 3.8 days, radon gas from natural sources can accumulate in buildings, particularly in confined or unventilated spaces.

Radon is estimated to cause between 3 % and 14 % of all lung cancer cases, depending on the average radon level in the country. For Europe, this corresponds to around 15 000 to 20 000 people per year dying of lung cancer caused by radon exposure.

Before the commencement of the EU-BSS, the recommended maximum limit for the annual ²²²Rn activity concentration at workplaces was 1 000 Bq/m³. With the new EU-BSS, for the first time, an obligatory maximum limit for the national legal limits of the ²²²Rn activity concentrations in indoor air of workplaces and dwellings was set for all EU member states.

Recently, the EU-BSS have been implemented into national legislation of the EU member states. The maximum reference limit set by the EU-BSS is 300 Bq/m³. A significant improvement in the metrological infrastructure in Europe for radon calibrations, especially at low activity concentrations (< 300 Bq/m³) was a prerequisite in order to be able to fulfil these requirements. Therefore, new procedures and traceable radon reference sources for the traceable calibration of radon measurement instruments at low activity concentrations with adequately low uncertainties needed to be developed.

Thoron (²²⁰Rn) and its progeny are known to bias the results of radon (²²²Rn) activity concentration measurements; however, information about this effect was limited. A better understanding of this effect was, therefore, needed together with techniques to reduce the influence of thoron and its progeny on radon measurements and calibrations. Traceability and quality assurance of calibrations of radon monitors and of radon calibration facilities, as well as the development of methods to conduct a large number of traceable and quality assured in-situ and laboratory measurements of radon were also required.

EU member states are bound to consider several aspects when preparing their national radon action plans (Article 103, EU-BSS), which is a strategy for conducting surveys of indoor radon concentrations. To ascertain that, the required level of safety is met for all European citizens, the consistency of indoor radon measurements and soil radon exhalation rate measurements across Europe needed to be optimised. Therefore, identification of Radon Priority Areas (RPA) was necessary in order to take appropriate actions for the protection of the public.

To reduce trade barriers and to ensure the mutual recognition of calibration certificates, general guidelines and recommendations on calibration and measurement procedures for the determination of radon concentration in air have to be established. This have been so far and will be in future facilitated too by the uptake of the project's results in the standards developing organisations and furthermore, by the end-users.

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research and innovation programme and the EMPIR Participating States

Publishable Summary



Objectives

The overall objective of this project was to enable the SI traceable monitoring of radon (²²²Rn) at low radon activity concentrations including calibration and radon mapping, in particular in support of the European Council Directive 2013/59/EURATOM (EU-BSS). The project contributed to the creation of a coordinated metrological infrastructure for radon monitoring in Europe.

The specific objectives of the project were:

- 1. To develop novel procedures for the traceable calibration of radon (222 Rn) measurement instruments at low activity concentrations (100 Bq/m³ to 300 Bq/m³) with relative uncertainties $\leq 5 \%$ (k = 1). As part of this, to develop new radioactive reference sources with stable and known radon emanation rates.
- 2. To investigate and to reduce the influence of thoron (²²⁰Rn) and its progeny on radon end-user measurements and radon calibrations.
- 3. To compare existing radon measurement procedures in different European countries and from the results optimise the consistency of indoor radon measurements and soil radon exhalation rate measurements across Europe.
- 4. To analyse and develop methodologies for the identification of Radon Priority Areas (i.e. areas with high radon concentrations in soil, as defined in the EU-BSS), including the development of the concept of a Radon Hazard Index (RHI), and to investigate the relationship between soil radon exhalation rates and indoor radon concentrations.
- 5. To validate traceability of European radon calibration facilities, and to publish guidelines and recommendations on calibration and measurement procedures for the determination of radon concentration in air.
- 6. To facilitate the take up of the technology and measurement infrastructure developed by the project by end users (regulators, radiological protection bodies and policy makers), standards developing organisations (ISO/TC45, CEN/TC351, ISO/TC85, CENELEC/TC 45, IAEA) and the measurement supply chain (accredited laboratories, instrumentation manufacturers).

Progress beyond the state of the art

In general, the technology and metrological infrastructure to assess the quality of radon measurements at low concentrations (< 300 Bq/m³) with sufficiently low measurement uncertainties was not available. Within this project, novel calibration methods and traceability validation at low radon activity concentrations have been devised and new and stable radioactive reference sources have been developed to enable these calibrations. Such calibrations in a stable radon atmosphere enable sufficiently low uncertainties to be achieved for low radon activity concentration measurements.

Although BIPM's Consultative Committee for Ionizing Radiation (CCRI(II)) comparisons of calibration for high radon activity concentrations (> 300 Bq/m³) had been conducted in the past, comparisons of existing radon gas standards at different European NMIs/DIs for ²²²Rn and ²²⁰Rn at low activity concentrations have been undertaken in this project.

For the first time, the distortion of the radon measurement results due to the presence of thoron has been considered and corrected at low radon activity concentrations within MetroRADON. For this purpose, reference thoron atmospheres had been established to calibrate secondary thoron reference instruments traceable to a primary thoron standard. Traceability to a primary thoron standard was ensured and refined enabling the thoron influence to be investigated. The influence of thoron on radon measurements have been studied using the traceably calibrated thoron reference instruments. Conclusions about the dependence of the radon monitor signal on the specific environmental conditions (radon to thoron ratio, temperature, time variations of radon/thoron concentrations and temperature) have been drawn together with the consequences for the design of radon surveys under real conditions.

The use of compact discs (CDs) and DVDs for retrospective radon measurements and their potential to define Radon Priority Areas have been evaluated. For this purpose, a long-time exposure experiment of CDs and DVDs had been set up. New techniques for measurement of radon exhalation from soil based on liquid



scintillation counting of polymers or track-etching of CDs have been developed and evaluated to analyse the results of the experiment.

The novel development of a European unified index of geogenic radon hazards, which has been defined flexibly independent of the data available, provides a consistent picture of susceptibility to geogenic radon across Europe. The novel definition of this Radon Hazard Index (RHI) is an important tool for the harmonised implementation and performance of national radon action plans of EU member states according to the EU-BSS requirements, which was not the case before. For this purpose, concepts and definitions of Radon Priority Areas (RPAs) which have been proposed and already implemented in the past and the role of stakeholders in the implementation process of RPA have been reviewed and evaluated together with approaches used to assess a "soil radon potential". Furthermore, existing mapping methods for RPAs used in various countries (e.g. indoor radon, gamma dose rate, geology, soil gas radon) have been tested.

The newly developed reference sources and procedures had been evaluated and compared to existing radon measurement procedures. Guidelines and recommendations on the new calibration and measurement procedures have been published based on the evaluation. Traceability of European radon calibration facilities using the new procedures and novel reference sources has been evaluated.

The project partners ensured that the results beyond the state of the art of this project have been so far and will be taken up in future by end users and standards organisations.

Results

Objective 1: Procedures for traceable calibration of radon (²²²Rn) instruments at low activity concentrations

Existing ²²²Rn emanation sources were re-evaluated concerning their present emanation power. Based on these investigations, prototypes of ²²²Rn emanation sources, which rely on electrodeposition, dropimplantation and chemisorption of radium (²²⁶Ra) were developed and produced. Additionally, a procedure for preparing ²²⁰Rn emanation sources was developed and tested successfully. Furthermore, a new low-level radon chamber has been designed, built, and tested to create long-time and time stable low-level radon atmospheres.

A long-term stable low-level ²²²Rn flow through source was designed, manufactured, and installed in two radon chambers. The source was tested at different air pressure, temperature, and humidity values. A combined standard measurement uncertainty of about 1 % (k = 1) could be achieved. Additionally, a method for the determination of activity concentration in a steady air flow has been developed and implemented. The feasibility of the method was tested successfully in air flows with ²²²Rn activity concentrations between 45 Bq/m³ and 100 000 Bq/m³.

Comparison protocols and schedules for the comparison of existing ²²²Rn gas primary standards and developed ²²⁰Rn emanation sources in the low kBq range were developed. ²²²Rn gas standards were produced and distributed to the participating laboratories. The results of this registered EURAMET supplementary comparison No. 1475, registered at BIPM as EURAMET.RI(II)-S8.Rn-222 were analysed and reported. All results are consistent, within the uncertainties. Five results are in very good agreement whereas two others are quite discrepant (about 20 % from the weighted mean). The relative standard deviation between the seven results is 11.4 %. The project successfully achieved the objective with these results.

Objective 2: Reducing the influence of thoron on radon measurements and calibrations

Three reference thoron atmospheres (low, intermediate, and high activity concentration) have been established at the reference chamber for calibration of measurement devices with the primary thoron standard system. The homogeneity of the reference atmospheres were tested by newly developed methods.

The calibrated measuring instruments are being used to study the influence of thoron on the radon measurement under field conditions and in two new exposure facilities to measure pure and mixed ²²⁰Rn and ²²²Rn atmospheres. Static and dynamic temperature conditions within -15 °C to + 60 °C have been created as well as predefined variable concentration profiles. A literature review of potential techniques and materials to reduce the influence of thoron on radon measurements provided the necessary data to start an experimental study on radon permeability and radon transport at different temperatures. A model for the evaluation of the performance of polymers as thoron barriers and a method based on differential measurements with two detectors for correction of the temperature bias have been developed and tested successfully.



Direct experimental determination of the radon transmission through polymer foils for different temperatures was performed. Radon transmission increases with temperature increase, while the detectors' response decreases. Compensating modules were designed in which detectors are placed, so that the thoron interference on the results is eliminated and both temperature bias and influence of humidity are greatly reduced. A patent for this technique was registered (SUBG: Bulg. Patent Application 112897/19.03.2019). A theoretical model for radon/thoron transmission that is usable for module design was developed.

IEC standard 61577 was reviewed and tested. It was found to be vague and not suitable for field measurements, as well as not covering most used detectors (e.g. SSNTDs). The findings had been merged and published as "Review of potential techniques and materials to reduce the influence of thoron on radon measurements and calibrations". The project successfully achieved the objective with these results.

Objective 3: Comparison of existing radon measurement procedures in different European countries and optimisation of the consistency of indoor radon measurements and soil radon exhalation rate measurements

An extensive literature review on existing indoor radon survey and geogenic radon surveys in Europe had been conducted. Two questionnaires, one on indoor radon surveys, and one on geogenic radon surveys conducted in Europe were prepared and sent to stakeholders. Although solid state nuclear track detectors were used in 82 % of the studies, the analysis of the results of the questionnaire on indoor radon surveys showed that the designs of surveys in Europe are not comparable. According to the questionnaire, 44 % of surveys were not performed during the whole year, and seasonal corrections were not applied in all of them. Seasonal variation of radon could differ within a country from region to region due to different factors, such as climate, living habits, building construction, etc. Although a large percentage of participants knew about the interference of thoron on radon measurements, they did not correct or check for thoron presence. A study of the representativeness of indoor radon measurement over different measurement periods (durations and seasons) was performed. Seasonal correction factors for estimating radon exposure in dwellings have been discussed.

Another literature review on the applicability of the CDs/DVDs for indoor radon surveys showed that CDs and DVDs can be used as ²²²Rn detectors in radon surveys. Existing standards EN ISO 11665-7 and ISO 11665-11, on the methodology of the radon exhalation measurement and of radon concentration in soil gas measurement, were successfully reviewed.

An intercomparison exercise based on variable indoor radon conditions along with ambient parameters located at the facilities of a former uranium mine near Salamanca, Spain, were conducted with 20 participants. Indoor radon gas measurements for passive and continuous radon monitors were carried out at around 300 kBq·m^{-3.}h and 2000 kBq·m^{-3.}h. Radon exhalations from soil and radon concentrations in soil gas were measured in the outdoor area. More than 80 % of the results for radon activity concentration in air were within the interval defined by the reference value and the standard deviation. The project successfully achieved the objective with these results.

Objective 4: Methodologies for the identification of Radon Priority Areas and investigation of the relationship between soil radon exhalation rates and indoor radon concentrations

An extensive literature review was conducted, reviewing, and evaluating the concepts and definitions of Radon Priority Areas proposed or already implemented. Methods developed in Europe to assess the "soil radon potential" which can be used for radon mapping and priority area definition were reviewed. Different approaches were selected to be tested and compared based on a case study from a region in France. Sources of RPA classification uncertainty were identified and ways to estimate and to quantify RPA uncertainty were proposed.

Data sets comprising the region of Cantabria (Spain) and six municipalities in Austria for testing existing radon mapping methods have been compiled and tested. The data include indoor radon measurements, building characteristics of measured dwellings, soil air radon activity concentrations, permeability, activity concentrations of soil, ambient dose rates, and maps of geogenic parameters. Differences between radon mapping and definition of RPAs across country borders were examined (Portugal / Spain, Spain / France, Switzerland / France, Belgium / France). To develop a complementary approach for mapping RPAs, data on geology, geochemistry, gamma radiation, indoor radon measurements and associated buildings characteristics at a national and local scale in regions with very high radon levels in Spain, Portugal and France were collected and validated. Statistical tools were applied to identify sectors with the highest indoor radon concentrations.



In addition, a long-term exposure experiment of CDs/DVDs as ²²²Rn detectors was conducted. It was found that the overall good correspondence between CDs/DVDs and the reference measurements even at low ²²²Rn activity concentrations (<100 Bq/m³) can be further improved if the correlation between radon concentrations and temperature during exposure is taken into account. A modified method, based on CDs/DVDs, can be used for measurement of low ²²²Rn concentrations, as well as for measurements in soil gas and for measurements determining the radon exhalation from soil surface. The project successfully achieved the objective with these results.

Objective 5: Validation of the traceability of European radon calibration facilities, and guidelines on calibration and measurement procedures for the determination of radon concentration in air

A survey of European calibration facilities, institutes with primary and secondary radon calibration standards, and selected end-users had been conducted. The results were evaluated, and 15 facilities were selected for validation and traceability exercises using an AlphaGUARD radon monitor as a reference instrument. The comparison exercise validated the traceability, performance, and precision of European radon calibration facilities in the range of 300 Bq/m³ to 10 000 Bq/m³ at three calibration points (400 Bq/m³, 1 000 Bq/m³ and 6 000 Bq/m³). Following the development of constant radon activity concentrations in reference chambers and calibration procedures at low activity concentrations, an intercomparison exercise to validate the traceability of the secondary standards used by the European radon calibration facilities was conducted.

Furthermore, the schedule and comparison protocol to validate the traceability of European radon calibration facilities at stable radon atmospheres in the range from 100 Bq/m³ to 300 Bq/m³ was developed. The project successfully achieved the objective with these results.

All results reported in detail are downloadable at http://metroradon.eu/index.php/documents/.

Impact

The project helped to establish a basic European metrological infrastructure for radon measurements enabling sound monitoring of radon and radon protection in Europe. Over the course of the project, about 300 European stakeholders and interested parties have been reached through the MetroRADON webpage, the newsletter, participation and organisation of workshops, presentations at scientific conferences and publications of peer-reviewed articles in scientific journals. The stakeholders consisted of manufacturers of radon monitoring equipment, companies offering radon measurements, calibration facilities, national authorities charged with the implementation of the EU Basic Safety Standards into national law, international bodies, etc. The stakeholders have been regularly updated with the project results.

The topicality of radon and the dissemination of the project's result created a strong interest in collaboration, resulting in nine academic and public organisations approaching the project to become involved as collaborators.

The project's results have been published in 15 peer reviewed scientific articles (six more papers have been drafted), used in seven technical workshops and disseminated in 70 presentations at European, international, and national conferences. The findings have been published in guidelines and best practice guides.

Impact on industrial and other user communities

This project has helped to establish a basic infrastructure for low radon activity concentration measurements and thus assist with the generation of Radon Hazard Index (RHI) maps so that metrologically sound measurements are possible to support the implementation of the EU-BSS and sound decision making for radiological protection and ensuring public health.

The impact of the project was firstly targeted to the process of the transposition of the EU-BSS, but beyond this, the project has enabled uptake and exploitation of its results and experience by all stakeholders concerned with radon, including the scientific community. As a consequence, field measurements in Europe have a higher precision, allowing more appropriate precautions and counter measures against public exposure to radon. In addition, harmonisation and standardisation has allowed the comparison and merging of different existing radon data sets on a European scale. The credibility of reported radon data has improved due to harmonised measurement procedures and has led to more consistent results. The relevant authorities and end users in EU member states have the possibility to perform accurate radon monitoring



due to improved calibration methods, summarised in best practice guides and recommendations on metrologically sound calibration and measurement procedures for the determination of radon concentration in air. A guideline summarizes the output of the project considering links between metrological standardization and radon mapping: <u>"Guideline on the constituents of the chain "from primary standards to radon maps" (traceability) and the links between them.</u>"

A large number of recommendations and guidelines, such as recommendations on the construction of radon monitors that are not sensitive to thoron have been developed and technical concepts/solutions aimed at reducing thoron-related bias to the radon signal in existing monitors have been published, thus leading to improved radon measurements and the opportunity for improved instrumentation.

An industry interest group, established in the project, provided a specialized platform to disseminate the projects results to stakeholders and pursue discussion and uptake. In June 2019, an Industry Interest Group (IIG) Meeting was achieved, and 20 representatives of highly relevant industry sectors attended. At this meeting, the demand of industry on radon monitoring has been discussed based on the presented aims and interim results of the present project.

Impact on the metrology and scientific communities

Stakeholders, regulators, and end-users of radon measurement methods profited from the dissemination of the results of this project. For this purpose, an industry interest group, group of national authorities and a group of European and international bodies and associations dealing with radon issues has been formed and used as channels to disseminate relevant information to different stakeholders. To inform all stakeholders of the on-going research conducted in MetroRADON, a project newsletter has been sent to approx. 300 recipients. The newsletter was facilitating the uptake of the procedures developed in the project by the endusers. To obtain input from stakeholders, questionnaires had been sent to competent institutions in Europe to obtain information about the primary and secondary standards used, the reference atmospheres, procedures and traceability chains, as well as on indoor radon surveys and geogenic radon surveys. The results of the surveys have been used for recommendations and guidelines that have been discussed in the scientific and metrological communities and published for end-users.

Ensuring traceability for the most commonly used European radon calibration facilities, especially at low radon activity concentrations, has significantly reduced calibration uncertainties. Impact was expected in the short term as various stakeholders have requested calibration services for instruments measuring the radon concentration in air. The metrological facilities for radon measurement instruments and detectors calibration improved and updated by the outcome of this project is summarized in the <u>Report "Network of European</u> calibration laboratories for radon concentration in air measurement".

The consideration of thoron in the newly developed calibration and measurement procedures triggered the development of new and improved methods in the scientific community and, therefore, the end-user community.

The project was facilitating the opportunity to conduct supplementary comparisons (EURAMET 1475 and BIPM/CCRI(II) as EURAMET.RI(II)-S8.Rn-222) for low radon activity concentrations. Through this, national metrology institutes (NMIs) and designated institutes (DIs) have had the ability to submit or expand their CMC entries for radon measurements. This enhanced the application of the mutual recognition arrangement (MRA) in that specific field of radionuclide metrology.

Impact on relevant standards

The project's outputs and data benefited European and international standards on radon monitoring and guidelines on radiological protection, construction products, radiation instrumentation and nuclear data. In particular, input has been given to:

- IEC TR 61577-5 "Radiation protection measurement instrumentation Radon and radon decay product measuring instruments – Part 5: General properties of radon and radon decay products and their measurement methods" (IEC TC 45/SC 45B Radiation protection instrumentation – WG 10: Radon and radon daughter measuring instruments),
- "Measurement of radioactivity in the environment Air: radon-222 QA/QC for calibration facilities" (ISO/TC 85/SC 2 Radiological protection WG 17 Radioactivity measurements), and
- Transformation of the CEN Technical Report TR 17113:2017 "Construction products: Assessment of release of dangerous substances Radiation from construction products Dose assessment of



emitted gamma radiation" into a CEN Technical Standard (EC Action on CEN TC 351 WG 3 Construction products: Assessment of release of dangerous substances, Radiation from construction products).

- Input to a EURAMET standard comparison report: EURAMET comparisons for ²²²Rn, not involving any Radon chambers, had been carried out about 15 years ago. The JRP comparison for ²²²Rn and ²²⁰Rn, done in WP1, has been successfully established as EURAMET TC IR comparison.
- Input to CENELEC/TC 45 Technical Report CLC/FprTR 62461 Radiation protection instrumentation Determination of uncertainty in measurement CLC/FprTR 62461:2019.
- Input to the revised IAEA Specific Safety Guide No. SSG-32 'Protection of the Public against Exposure Indoors due to Radon and Other Natural Sources of Radiation', jointly sponsored by the IAEA, WHO. BEV and other JRP partners support the IAEA radon section.
- Input to ICRM guideline on radon metrology. BEV and other JRP partners presented the progress and the interim results of the JRP in the ICRM Low Level Radionuclide Metrology Techniques Working Group at the 22nd International Conference on Radionuclide Metrology and its Applications, at the University of Salamanca (May 2019).

This input has improved the traceability requirements of radon and radon daughter measuring instruments in European and international standards. The aims of which are the reduction of uncertainties at instrument calibrations and harmonization of calibration methods at low radon activity concentrations (< 300 Bq/m³ ²²²Rn). Input has also been given to IEC standard 61577 at the end of the project.

Longer-term economic, social, and environmental impacts

In Europe thousands of lung cancer cases annually are attributed to indoor radon progeny exposure. This project contributed to improving public health through more reliable radon measurements as a basis for effective radon mitigation and prevention of radon progeny induced lung cancer in Europe, leading to improved public health and reduced healthcare costs.

List of peer-reviewed international journal publications

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- K. Mitev , P. Cassette, D. Pressyanov, S. Georgiev, Ch. Dutsov, N. Michielsen, B. Sabot. Methods for the experimental study of ²²⁰Rn homogeneity in calibration chambers. Applied Radiation and Isotopes. <u>https://zenodo.org/record/4160815</u>
- 4. Georiev, S., Mitev, K. et al. Partition coefficients and diffusion lengths of radon in some polymers at different temperatures. International Journal of Environmental Research and Public Health. https://doi.org/10.3390/ijerph16224523
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https://zenodo.org/record/3555047#.YFItSZ37RPY; https://doi.org/10.1016/j.apradiso.2019.108907 publisher

link

- Sahagia M. Education and training tradition at IFIN-HH in radon measurement and evaluation of radiological impact. Romanian Reports in Physics 71, 906 (2019). <u>http://www.rrp.infim.ro/2019/AN71906.pdf</u>
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- 11. Bossew, P., Radon Priority Areas Definition, Estimation and Uncertainty, Nuclear Technology and Radiation Protection 33, 3 (2018) p. 286-292, <u>https://doi.org/10.2298/NTRP180515011B</u>
- 12. Bossew, P., Radon Priority Areas and Radon Extremes Initial Statistical Considerations, Radiation Environment and Medicine 8, 2 (2019), p. 94–104, <u>http://crss.hirosaki-u.ac.jp/wp-</u> <u>content/files_mf/1568795052Web_REMVol828_PeterBossew.pdf?wb48617274=17147ED8</u>
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This list is also available here: <u>https://www.euramet.org/repository/research-publications-repository-link/</u>

Project start date and duration:		1 June 2017, 42 months	
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1 BEV-PTP, Austria	9 AGES, Austria		17 METAS, Switzerland
2 BFKH, Hungary	10 BfS, Germany		
3 CEA, France	11 CLOR, Poland		
4 CMI, Czech Republic	12 IRSN, France		
5 IFIN-HH, Romania	13 JRC, European Commission		
6 PTB, Germany	14 SUBG, Bulgaria		
7 STUK, Finland	15 SUJCHBO, Czec	h Republic	
8 VINS, Serbia	16 UC, Spain		
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