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Radioxenon standards used in laboratory inter-comparisons

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HIGHLIGHTS

- Development of two independent ^{133}Xe activity concentration standards.
- Traceability to international reference standards established.
- Good agreement of the two ^{133}Xe activity concentration standards.
- Xe-127 shows to be a suitable isotope for QA/QC of remote IMS noble gas systems.

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ABSTRACT

Preparation methods for ^{133}Xe standards of activity concentration and the results of the 2014 ^{133}Xe laboratory inter-comparison exercise are described. One element of the quality assurance/quality control (QA/QC) program for laboratories of the International Monitoring System (IMS) will be regular inter-comparison exercises. However, until recently, no activity concentration standards for benchmarking were available. Therefore, two ^{133}Xe activity concentration reference standards were produced independently by Idaho National Laboratory and Seibersdorf Laboratories and used for the 2014 laboratory inter-comparison exercise. The preparation of a complementary ^{127}Xe activity concentration standard as well as a ^{127}Xe laboratory inter-comparison exercise suggests ^{127}Xe as a suitable isotope for QA/QC of remote IMS noble gas stations.

1. Introduction

As part of the verification for Comprehensive Nuclear–Test–Ban Treaty (CTBT) compliance, the Provisional Technical Secretariat (PTS) of the CTBTO Preparatory Commission, together with Member States, is establishing an international monitoring network. When completed, the International Monitoring System (IMS) will consist of 321 monitoring stations comprised of four technologies: seismic, infrasound, hydroacoustic and radionuclide monitoring. The radionuclide component of the IMS will consist of 80 radionuclide stations, of which 63 have already been installed and are certified according to IMS station requirements (CTBT/PTS/INF.58/Rev.8, 2007). All radionuclide monitoring stations will be equipped with air samplers and high purity germanium detectors to detect and measure aerosol borne (particulate) radionuclides. The network of radionuclide stations is supported by 16 laboratories, certified according to IMS radionuclide laboratories requirements, for quality assurance and corroboration of station results. (CTBT/PTS/INF.96/Rev.9, 2012).

Particularly for the detection of underground or underwater nuclear explosions, at least forty of the eighty radionuclide stations will also be equipped with systems to measure the radioactive noble gas isotopes ^{131m}Xe ($T_{½}$=11.962 (20) d), ^{133}Xe ($T_{½}$=5.2474 (5) d), ^{133m}Xe ($T_{½}$=2.198 (13) d) and ^{135}Xe ($T_{½}$=9.14 (2) h). All nuclear data was taken from the DDEP project (http://www.nucleide.org/DDEP_WG/DDEPdata.htm). These are among the radioisotopes with the highest yields in fission of uranium or plutonium. In underground nuclear explosions, noble gases can

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escape into the atmosphere, while most of the other fission products (the particulates) condense quickly after explosion and are usually trapped in the surroundings of the test cavity (e.g. Carrigan et al. (1996), De Geer (1996)). Similarly, in underwater explosions, noble gases have a higher chance to be vented into the atmosphere than particulates. Currently, 22 out of the forty noble gas detection systems have been certified according to IMS noble gas station requirements (CTBT/PTS/INF.921/Rev.3, 2008).

2. Measurement technology

When the establishment of the IMS radionuclide network commenced in 1997, there was only limited experience with noble gas monitoring and, until the mid-1990s, only laboratory-based manual systems were available. In the course of the IMS development and set-up, automated systems for noble gas monitoring, suitable for operation at remote sites, were developed. In current systems, the sampling process of noble gases is mostly based on the adsorption of xenon on activated charcoal. After sampling, the xenon is desorbed from charcoal by heating and flushing with a carrier gas (either nitrogen or helium), which is further purified with the aim to obtain a sample of xenon and carrier gas only. The determination of xenon activity concentrations in air is done in a two-step procedure – determination of the sampled air volume and of the xenon activity extractable from that volume.

The measurement of the air volume of the sample is done by quantifying the volume of xenon gas in the counting cell. Natural occurring xenon is present in atmospheric air at an abundance of 87 parts per billion by volume (ppbv), as defined in the U.S. Standard Atmosphere (CRC2014, 2014). Therefore, the corresponding air volume of a sample, V_{air}, is determined by $V_{\text{air}} = V_{\text{Xe}}/0.087*10^{-6}$, where V_{Xe} is the volume of xenon in the sample. As a consequence, the activity concentration, $<A>$, of a sample is:

$$<A> = \frac{\textit{Sample Activity}\ [\text{Bq}]}{\textit{Xenon Volume}\ [\text{cm}^3]} * 0.087 \frac{[\text{cm}^3]}{[\text{m}^3]}$$

The xenon volume measurement is done with a thermal conductivity detector either in a static volume or in a gas chromatograph (Ringbom et al., 2003; Fontaine et al., 2004; Dubasov et al., 2005).

Activity measurements with IMS noble gas (NG) systems are done either by high-resolution gamma-ray spectrometry or beta-gamma coincidence counting. One requirement for the NG systems deployed in the IMS is to have a minimum detectable concentration for ^{133}Xe of $\leq 1\ \text{mBq/m}^3$as defined in (CTBT/PTS/INF.921/Rev.3, 2008); the total sample air volume is typically 10–75 m^3, depending on the type of noble gas system (Auer et al., 2010).

3. QA/QC requirements

To ensure a high quality of data provided by the IMS NG systems, a comprehensive quality assurance/quality control (QA/QC) program needs to be established. One component of such a QA/QC program is the evaluation of station performance through independent measurements of samples from the deployed systems by participating laboratories. For this purpose, either routine ambient air samples or samples "spiked" with radioactive xenon can be used. The actual parameters measured at noble gas stations and by laboratories are the activity of xenon isotopes and the volume of xenon gas in the sample. In general, a perfect match of values measured at the noble gas station and at the laboratory it is not expected, because of a potential loss of gas or dead volumes not accounted for by the noble gas systems.

Xenon activity concentrations and isotope ratios are used as key parameters because they are independent of any gas losses. A deviation of station and lab result of these parameters within 15% is generally considered acceptable. Samples containing both ^{131m}Xe and ^{133}Xe are the most suitable, owing to sufficiently long half-lives of these two isotopes to account for the transportation time between stations and laboratories. However, routine re-measurement of ^{133m}Xe is quite challenging, and not feasible for ^{135}Xe due to short half-life.

Laboratory measurements are crucial in a QA/QC program. As a consequence, good accuracy and traceability of the actual measurements are also of critical importance in order to provide confidence in the results. Therefore, in order to ensure the credibility of IMS laboratories as providers of reference results, the QA/QC program for laboratories must include regular proficiency test exercises (PTE) to assess the performance of a laboratory against pre-established criteria.

Currently six IMS radionuclide laboratories have noble gas measurement capabilities (using in-house developments and commercially available equipment) for the four CTBT-relevant xenon isotopes (^{131m}Xe, ^{133}Xe, ^{133m}Xe and ^{135}Xe) and are able to perform on-request reanalysis of NG samples from the certified IMS NG stations. These laboratories have participated in a QA/QC pilot program for noble gas measurements.

4. Xenon inter-comparison exercises

Several laboratory inter-comparison exercises have been conducted since 2008 (Gohla et al., 2011; Gohla and Auer 2013), initially with only four laboratories participating (three IMS laboratories and one non-IMS laboratory). The purpose of these exercises were to assist the laboratories in the development of noble gas measurement capabilities and to eventually establish regular proficiency test exercises as part of the QA/QC program. To that end, nearly identical samples (or one sample split for distribution among the participants) were sent to the participating laboratories by the producers. Quality checks (e.g., leak testing of sample containers, xenon homogeneity) were conducted. Sample transport from the producer to the laboratories was usually completed within five days.

Although international comparisons of ^{133}Xe activity standards exist (Ratel and Michotte 2004), no activity concentration standards traceable to the SI were available. Consequently, for previous (2008–2012) inter-comparisons among laboratories, no references were available for benchmarking.

Fig. 1 shows a typical result of an inter-comparison exercise of

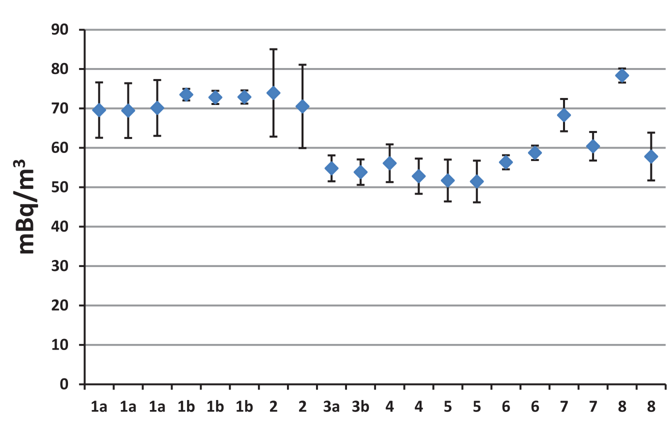

Fig. 1. ^{133}Xe activity concentration results of an inter-comparison exercise in 2012. Uncertainties are shown for $k = 1$.