Applied Radiation and Isotopes 103 (2015) 102–114

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Spalax™ new generation: A sensitive and selective noble gas system for nuclear explosion monitoring

G. Le Petit*, A. Cagniant, P. Gross, G. Douysset, S. Topin, J.P. Fontaine, T. Taffary, C. Moulin

CEA, DAM, DIF, F-91297 Arpajon, France

HIGHLIGHTS

- CEA developed a new noble gas station (SPALAX™ NG) for Comprehensive Test Ban Treaty purposes.
- SPALAX™ NG is able to extract more than 6 cm^3 of pure xenon in 70 m^3 air sampled during 12 h.
- Atmospheric radioxenon are measured using HPGe/Si electron-photon coincidences.
- SPALAX™ NG Minimum Detectable activity Concentrations are about 0.1 mBq/m^3 for $^{133,133m\ and\ 131m}Xe$ and 0.4 mBq/m^3 for ^{135}Xe.

ARTICLE INFO

Article history:
Received 22 October 2014
Received in revised form
14 April 2015
Accepted 27 May 2015
Available online 28 May 2015

Keywords:
Spalax™
Pipsbox™
Electron-photon coincidences
Radioxenon
Nuclear explosion monitoring

ABSTRACT

In the context of the verification regime of the Comprehensive nuclear Test ban Treaty (CTBT), CEA is developing a new generation (NG) of SPALAX™ system for atmospheric radioxenon monitoring. These systems are able to extract more than 6 cm^3 of pure xenon from air samples each 12 h and to measure the four relevant xenon radioactive isotopes using a high resolution detection system operating in electron–photon coincidence mode. This paper presents the performances of the SPALAX™ NG prototype in operation at Bruyères-le-Châtel CEA centre, integrating the most recent CEA developments. It especially focuses on an innovative detection system made up of a gas cell equipped with two face-to-face silicon detectors associated to one or two germanium detectors. Minimum Detectable activity Concentrations (MDCs) of environmental samples were calculated to be approximately 0.1 mBq/m^3 for the isotopes ^{131m}Xe, ^{133m}Xe, ^{133}Xe and 0.4 mBq/m^3 for ^{135}Xe (single germanium configuration). The detection system might be used to simultaneously measure particulate and noble gas samples from the CTBT International Monitoring System (IMS). That possibility could lead to new capacities for particulate measurements by allowing electron–photon coincidence detection of certain fission products.

1. Introduction

Comprehensive nuclear Test-Ban Treaty (CTBT) verification regime opened for signature within the United Nations General Assembly (UNGA) in 1996 is based on a world-wide network of sensors: the International Monitoring System (IMS). It is based on four different technologies: waveform sensors (seismic, hydro-acoustic and infrasound), radionuclide sensors (global monitoring of radioactive aerosols and radioactive noble gas) together with atmospheric transport modelling linked to the International Data Centre (IDC) to process and review all data provided by the sensors. In case of a low energy (in the order of kt (kiloton of equivalent TNT) or less) underground nuclear explosion, the detection of radioactive noble gases is likely to provide unambiguous evidence of a violation of the CTBT.

Among fission products under gaseous form created by a nuclear test, four relevant CTBT radioactive xenon isotopes (^{131m}Xe $T_{1/2}$= 11.93 d, ^{133m}Xe $T_{1/2}$= 2.19 d, ^{133}Xe $T_{1/2}$= 5.24 d and ^{135}Xe $T_{1/2}$= 9.14 h) are of interest. These isotopes are produced in significant amounts as they are in the mass range 131–135, near the maximum of the fission product mass-yield of both uranium and plutonium. Furthermore, these radioxenons have half-lives long enough to be detected at significant distances even several days or weeks after the initial explosion.

When an underground nuclear explosion occurs, owing to both their inert chemical properties and ability to be mobile, radioactive noble gas at trace level can escape from natural faults, cracks and fractures produced by the detonation and reach the surface. Even in the case of a well-contained underground nuclear explosion, highly diluted radioactive gases have been shown to migrate to surface within periods of weeks to several months due to natural gas transport processes driven by barometric pressure

* Corresponding author. Fax: +33 1 69 26 70 65.
E-mail address: gilbert.le-petit@cea.fr (G. Le Petit).

http://dx.doi.org/10.1016/j.apradiso.2015.05.019
0969-8043/

oscillation (Carrigan et al., 1996). Another possibility for noble gas release within the atmosphere can be due to human intervention within the tunnel some weeks after the nuclear test for diagnostic purposes. A striking case illustrating the high capability of the IMS noble gas network to detect delayed signals from a nuclear explosion is given by the radioxenon isotopic ratios $^{131m}Xe/^{133}Xe$ measured in April 2013 at the Takasaki IMS station (Japan). These signals based on atmospheric transport conditions prevailing at that time and on the study of the station detection records, were considered to be possibly linked to releases of radioactive xenon 7–8 weeks following the nuclear test conducted on February 12, 2013 by North Korea (Ringbom et al., 2014).

Four systems capable of sampling, processing and analysing airborne radioxenons have been developed for the CTBT verification regime: the Swedish Automatic Unit for Noble gas Acquisition (SAUNA, Ringbom et al., 2003), the Automated Radioxenon Sampler and Analyser (ARSA, Bowyer et al., 1996), the Analyser of Xenon RadioIsotopes (ARIX, Dubasov et al., 2002) and the Système de Prélèvement Automatique en Ligne avec l'Analyse du Xénon atmosphérique (SPALAX™, Fontaine et al., 2004). These systems perform radioxenon detection either using high–resolution HPGe gamma spectroscopy (SPALAX) or $\beta-\gamma$ coincidence spectroscopy (ARSA, ARIX and SAUNA). Minimum Detectable activity Concentrations (MDCs) for isotopes ^{133}Xe and ^{135}Xe are well under the CTBT requirement (<1 mBq/m^3 for the isotope ^{133}Xe) and quite similar (0.2–0.3 mBq/m^3 for ^{133}Xe) whatever the system. Better MDCs for metastable isotopes (^{131m}Xe and ^{133m}Xe) are achieved using $\beta-\gamma$ detection systems (<1 mBq/m^3). However, owing to their low energy resolutions, both for electrons (measured by a plastic scintillator detector) and photons (measured by a NaI(Tl) detector) (Mekarski et al., 2009), these systems need to use robust algorithms (Axelson et al., 2003, MacIntyre et al., 2006) to solve interferences in the coincidence region of interest (ROI). A high resolution photon/electron coincidence system requires fewer corrections than low resolution systems regarding interference between radio-isotopes eventually present in a sample. This results in lower uncertainties on the measured activities.

Another point that came to light in the early 2000's is related to the increase of civilian radioactive sources (Saey, 2009). Hence, main airborne radioxenon contributors are due to operations of nuclear power plants (NPPs), and accidental or controlled releases from medical isotope production facilities (IPFs). These radioxenon emission sources come from the production of fission based ^{99}Mo, which decays to ^{99m}Tc, from uranium targets irradiated in reactor. Technetium 99m is broadly used in disease diagnosis and therapies for approximately 30 million treatments per year. Consequently, atmospheric release levels from fission-based ^{99}Mo production, varying within the range 10^9–10^{13} Bq per day, can lead to radioxenon backgrounds likely to mask a nuclear explosion signal or unlike NPPs to bias isotopic ratio information and reduce confidence on a relevant nuclear test signature (Bowyer et al., 2013, 2014) especially at low level detection.

In parallel to the implementation by the major medical radioisotope producers of new technologies to mitigate radioxenon emissions, efforts to maintain the capability of the CTBT verification regime to show clear evidence of a violation of the nuclear treaty are undertaken. A promising way is to design a new generation of noble gas monitoring systems: more sensitive and accurate compared to the current IMS ones together with shorter air sampling time. These new systems should significantly improve the sampling volume while keeping an effective xenon extraction processing and by drastically improving performances of the detection system. In particular, the detection technologies associated to these new xenon monitoring systems have to lead to weaker spectral interference correction levels in order to provide more accurate measurements. Consequently, lower uncertainties in computed ratios would allow a better identification of the mixing of multiple source signatures likely to increase confidence on a potential relevant signature. Recent works on detection systems (Alemaychu et al., 2014, Xie et al., 2013, Hennig et al., 2013) based on new technologies are providing promising results.

This work addresses a new generation of noble gas systems (SPALAX™ NG) operating at CEA/DAM Ile-de France, near Paris (France), able to process more than 6 cm^3 of pure xenon (70 m^3 air equivalent) from 12 h air sampling and especially focused on an innovative high energy resolution detection system using electron–photon coincidence technique in order to analyse the four relevant CTBT radioactive xenon isotopes at very low activity concentrations. It is shown that the detection geometry of this new spectrometer allows for a simultaneous measurement of the two types of relevant CTBT samples, respectively a particulate filter and a noble gas sample, without significantly affecting either spectral information. This new insight into the CTBT verification regime field might be conceivable for a next generation of IMS detection systems in the context of the technology foresight as discussed in the last section of this article.

2. Spalax™ NG concepts

The SPALAX™ NG relies on the most recent CEA researches and developments to reach the maximum radioactive noble gas detection sensitivity and selectivity together with high reliability. For that purpose, R & D has focussed on major improvements and/or breakthroughs on the three major components of the system automatically controlled via a programme logic controller (PLC): the air sampling, the air processing and the detection system. The main objective of this programme was to design a noble gas system providing 2 samples per day with better MDCs than those reached by the current IMS noble gas systems.

The air sampling unit is significantly improved by replacing the current SPALAX™ compressor technology (scroll pump) by a dry piston pump or by a screw pump. Both technologies enable to increase the sampling flow rate by at least a factor 2 while keeping energy consumption almost constant. Both technologies are still under long term reliability tests but have already exhibited a higher lifetime than the current scroll pump. Concerning the gas processing, the number of nitrogen membranes is adjusted to the new flow rate enhancing xenon concentration into nitrogen at least of a factor 2 while depleting in oxygen and in carbon dioxide. The new sampling unit is able to produce a gas up to 3 ppm Xe while the current SPALAX™ produces 1 ppm Xe (Fontaine et al., 2004). The air processing unit includes also a major evolution within the concentration/purification process. Hence, a silver doped zeolite (MFI zeolite) has been tested (Daniel et al., 2013; Deliere, 2015) at the different stages of the process in place of the current carbon adsorbent materials, activated charcoal for the ovens dedicated to the first concentration and purification steps and carbon molecular sieve for those related to the ultimate concentration step. A major increase of xenon retention capacity, by at least a factor 10 compared to the current activated charcoal, has been achieved. The material is so efficient that it allows keeping only one single column for the last concentration step. These major evolutions lead to improve the overall SPALAX™ process efficiency and to reduce its size by approximately 20% while lowering energy consumption by a factor 2. Long term stability of the new adsorbent material is still in progress. In sum up, the new SPALAX™ process is able in 12 h, to recover almost 6–7 cm^3 of xenon (up to 70–80 m^3 of equivalent air volume) compared to the 2–3 cm^3 of xenon for the current SPALAX™ systems. At the end of the process, the last sample production step has to be optimized in order to efficiently recover the new xenon volume