

Ten Years of Development of Equipment for Measurement of Atmospheric Radioactive Xenon for the Verification of the CTBT

MATTHIAS AUER,¹ TIMO KUMBERG,¹ HARTMUT SARTORIUS,¹ BERND WERNSPERGER,² and CLEMENS SCHLOSSER¹

Abstract—Atmospheric measurement of radioactive xenon isotopes (radioxenon) plays a key role in remote monitoring of nuclear explosions, since it has a high capability to capture radioactive debris for a wide range of explosion scenarios. It is therefore a powerful tool in providing evidence for nuclear testing, and is one of the key components of the verification regime of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). The reliability of this method is largely based on a well-developed measurement technology. In the 1990s, with the prospect of the build-up of a monitoring network for the CTBT, new development of radioxenon equipment started. This article summarizes the physical and technical principles upon which the radioxenon technology is based and the advances the technology has undergone during the last 10 years. In contrast to previously used equipment, which was manually operated, the new generation of radioxenon monitoring equipment is designed for automated and continuous operation in remote field locations. Also the analytical capabilities of the equipment were strongly enhanced. Minimum detectable concentrations of the recently developed systems are well below 1 mBq/m³ for the key nuclide ¹³³Xe for sampling periods between 8 and 24 h. All the systems described here are also able to separately measure with low detection limits the radioxenon isotopes ^{131m}Xe, ^{133m}Xe and ¹³⁵Xe, which are also relevant for the detection of nuclear tests. The equipment has been extensively tested during recent years by operation in a laboratory environment and in field locations, by performing comparison measurements with laboratory type equipment and by parallel operation. These tests demonstrate that the equipment has reached a sufficiently high technical standard for deployment in the global CTBT verification regime.

Key words: Comprehensive Nuclear-Test-Ban Treaty (CTBT), International Monitoring System (IMS), environmental monitoring, noble gas, radioxenon.

1. Radioxenon Measurements for Monitoring of Nuclear Tests

After many years of negotiation, the vote for the Comprehensive Nuclear-Test-Ban Treaty (CTBT) by the General Assembly of the United Nations on 10 September, 1996 marked a major milestone in nuclear nonproliferation (RAMAKER *et al.*, 2003; HANSEN, 2006). This treaty obliges its member states “not to carry out any nuclear weapon test explosion or any other nuclear explosion, and to prohibit and prevent any such nuclear explosion at any place under its jurisdiction of control” [Text of the CTBT, Article 1]. The implementation of this treaty is largely based on a verification system for monitoring nuclear tests underground, underwater and in the atmosphere in order to minimize the chance of clandestine testing. A crucial component of this verification regime is the International Monitoring System (IMS) which consists of 321 facilities worldwide and, once completed, will provide an unprecedented global coverage of facilities to monitor nuclear tests. The IMS is based on the combination and the synergy of facilities for seismic (BARRIENTOS and HASLINGER, 2001), hydro-acoustic (LAWRENCE *et al.*, 2001), infrasound (CHRISTIE *et al.*, 2001) and radionuclide monitoring (MATTHEWS and SCHULZE, 2001). In order to fulfil the ambitious task of the IMS in an optimized manner, state-of-the-art equipment is deployed at IMS stations. Therefore, with the onset of the CTBT, development of new equipment was triggered in all four monitoring technologies. The radionuclide component of the IMS consists of eighty facilities for monitoring aerosol-bound radioactive fission products (denoted as particulates), of which forty facilities are planned to be

¹ Federal Office for Radiation Protection, Rosastr. 9, 79098 Freiburg, Germany. E-mail: cschlosser@bfs.de

² Provisional Technical Secretariat of the Comprehensive Nuclear-Test-Ban Treaty Organization, 1400 Vienna, Austria.

equipped for monitoring radioactive xenon isotopes (denoted as radioxenon or noble gas) (see Fig. 1) and of 16 radionuclide laboratories, which provide further analysis of samples from the stations (KARHU and CLAWSON, 2001). At the first annual conference of state parties of the CTBTO, which will take place after entry into force of the CTBT (Text of the CTBT, Protocol, Part I, C, p. 130), the state parties will decide whether noble gas systems will be deployed throughout the IMS radionuclide network. This article focuses on the technical developments and advances in monitoring of radioactive xenon isotopes, which have been achieved during the last approximately 10 years and presents results that recommend a strong xenon role for monitoring the atmosphere.

Measurement of the radioactive xenon isotopes ^{131m}Xe ($T_{1/2} = 11.84$ days), ^{133}Xe ($T_{1/2} = 5.24$ days), ^{133m}Xe ($T_{1/2} = 2.19$ days) and ^{135}Xe ($T_{1/2} = 9.14$ h) (hereafter termed radioxenon) in the atmosphere is a crucial component of the IMS in particular for the detection of clandestine and underground tests (CARRIGAN *et al.*, 1996; PERKINS and CASEY, 1996; DE GEER, 1996a): Release of fission products other than noble gases from an underground test is unlikely and restricted to accidentally venting tests or operational releases (BJURMAN *et al.*, 1990), while for noble gases,

which are chemically inert, there is a considerably higher chance to be released through cracks and fissures in the ground. Xenon isotopes have the highest fission yield among noble gases with cumulative fission yields of 6–7% for ^{133}Xe , for a ^{235}U or ^{239}Pu fission device, respectively (ENGLAND and RIDER, 1993). This makes them the first choice as indicators for underground nuclear explosions. In addition, the only sink for radioxenon in the atmosphere is its radioactive decay, hence, the atmospheric residence time is equal to the mean radioactive lifetime, thus allowing for long-range transport in the atmosphere. Also, the typical atmospheric background level is low, e.g., in the order of 1 mBq/m^3 in Central Europe, Japan and US (DE GEER, 1996b, WEISS *et al.*, 1997; BOWYER *et al.*, 1997, 2002; IGARASHI *et al.*, 2000), due to lack of significant natural sources and due to the relatively short half-lives of the radioxenon isotopes. In contrast to radioxenon, the high atmospheric background of ^{85}Kr , currently approximately 1.5 Bq/m^3 in the Northern Hemisphere (HIROTA *et al.*, 2004) with a high temporal and geographical variability (WEISS *et al.*, 1992; WINGER *et al.*, 2005), makes the atmospheric concentration of this isotope relatively insensitive to emissions of nuclear tests. In addition, the fission yield of this isotope (cumulative

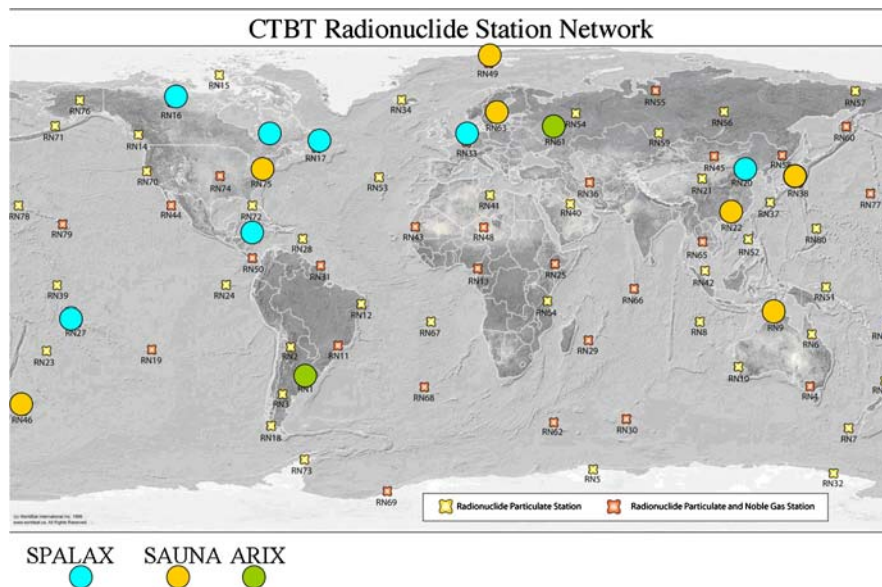


Figure 1

Radionuclide station network of the International Monitoring System. Blue, yellow and green dots indicate the system types at noble stations installed by September 2008 (Source: CTBTO IMS-Engineering and Development Section)

yield 0.2–0.4% (ENGLAND and RIDER, 1993) is approximately a factor 20 lower than the yield of, e.g., ^{133}Xe . Note that the fission yield for isobars of mass 85 is relatively high (approximately 1.5%), the low cumulative yield of ^{85}Kr is highly, because its precursor $^{85\text{m}}\text{Kr}$ beta-decays with a branching ratio of 79% to ^{85}Rb . The only other noble gas isotope, which aside from the xenon isotopes may be a sensitive indicator for an underground nuclear explosion is ^{37}Ar ($T_{1/2} = 35$ days) (LOOSLI, 1992). This isotope has a negligible natural and a very low man-made background on the order of 1 mBq/m^3 in the atmosphere (LOOSLI, 1992) and approximately up to two orders of magnitude higher in soil gas (Purtschert, pers. comm.). ^{37}Ar is produced in underground nuclear explosions by neutron activation of ^{40}Ca contained in the rock surrounding the explosion. Measurement of ^{37}Ar is however technically challenging. Firstly, because the energy of the Auger electrons emitted from the electron capture decay of ^{37}Ar is low (2.82 keV) and secondly, the high Ar content in the air results in a large sample size. Currently there is only one laboratory worldwide where ^{37}Ar is measured routinely in environmental samples, which is the low-level underground laboratory of the University of Bern (LOOSLI *et al.*, 1986). With the present available technology, ^{37}Ar can however, not be measured continuously in field-operated stations, although it is a very valuable and feasible technique for on-site inspections, where selective samples are taken close to the site of a presumed underground nuclear explosion.

What makes the measurement of radioxenon, and radionuclide measurements in general, an important and even indispensable component of the IMS is its unique capability to identify such a signal as originating from a nuclear explosion by measurement of the fission products. Using information on atmospheric transport processes, the signal measured by a radionuclide station can be attributed to a potential source region (WOTAWA *et al.*, 2003). The localisation is also supported by taking into account information from seismic, hydro-acoustic or infrasound measurements. Measurement of radioxenon has a long history in the monitoring of nuclear activities. Already during the Second World War, the United States conducted over-flights in Germany in 1944 to

search for nuclear reactors (ZIEGLER and JACOBSON, 1995). Since then, measurements of radioxenon have been widely used to monitor the release of fission products either from nuclear explosions or from civil nuclear activities (e.g., SCHÖLCH *et al.*, 1966; IGARASHI *et al.*, 2000). A recent example for the importance of radioxenon may be the test conducted by the Democratic Peoples Republic of North Korea on 9 October, 2006. To date the most convincing evidence for the nuclear character of the explosion is the detection of radioxenon by different groups (SAEY *et al.*, 2007; RINGBOM *et al.*, 2009; BECKER *et al.*, 2010).

2. General Requirements for IMS Noble Gas Equipment

Previous radioxenon measurement technology was based on manual analysis systems which were operated in a typical laboratory environment (EHHALT *et al.*, 1963; SCHÖLCH *et al.*, 1966; LUDWICK, 1966; STOCKBURGER *et al.*, 1977; BERNSTRÖM and DE GEER, 1983; KUNTZ, 1989). With the recent development of the IMS, new requirements for monitoring equipment have been set. The noble gas component of the IMS is based on the principle of a globally uniform monitoring coverage at a high time resolution (the sampling time is equal or less than 24 h) and a high sensitivity of the measurement systems (detection sensitivity for ^{133}Xe less or equal to 1 mBq/m^3). The high time resolution is of particular importance first, in order to constrain the time window for the atmospheric transport calculations for source location and second, in order to reduce the chance of interference with background, e.g., from civil sources. Stations are also often at remote sites with very limited infrastructure and access. This requires a high technical standard and reliability of the equipment in order to meet the stringent requirements for IMS stations. In addition, manual operation of radioxenon equipment requires a high level of technical expertise, which is not necessarily available at IMS sites and which would involve extensive and costly training of operators. Automated systems are therefore a more suitable choice for IMS sites than manual systems. The previously used and laboratory-based equipment

was not designed for such needs. This equipment usually required a high degree of manual operation by a system expert, which allowed the operator to interactively perform the sample analysis in order to adjust the analysis to the specific needs of the sample and optimize the results. This is not possible for the IMS, where a large amount of data has to be produced (40 noble gas stations with currently up to two samples per day). The data must be of uniform quality and quickly available at the International Data Center (IDC). The time allowed for reporting from the station to the IDC is 48 h after start of sampling.

In addition to these new operational challenges, the task to monitor nuclear tests, distinct from other nuclear activities, also sets very specific requirements for the analytical capabilities of the equipment. In particular for remote monitoring, high sensitivity of the equipment is required. Assuming venting of only independently produced ^{133}Xe , a 1 kT underground nuclear explosion may release in the order of 10^{14} Bq ^{133}Xe into the atmosphere. The order of distance from any possible source location on Earth to a radionuclide station is given by the typical distance between IMS radionuclide stations, which is approximately 1,000 km. Within this distance, the debris released from a nuclear explosion is diluted by a factor 10^{-14} to 10^{-18} (A. Becker, pers. comm.). Therefore, with a detection limit of 1 mBq/m^3 for ^{133}Xe , a release from a 1 kT nuclear explosion is likely to be detected by the IMS noble network (CD/NTB/WP.224, 1995).

The strength of a radioxenon signal alone is for many sampling locations an insufficient criterion, since civil sources may cause ambient concentrations of a few mBq/m^3 with short spikes of 10^2 to 10^3 mBq/m^3 , in particular in areas with a high density of nuclear facilities, such as Northern Europe, Asia or Northern America (e.g., AUER *et al.*, 2004; STOCKI *et al.*, 2005). The distinction can only be made by the isotopic composition, which is usually different for nuclear explosions and other nuclear activities (KALINOWSKI *et al.*, 2010) and by taking into account the characteristic concentrations and their typical variation at a specific site. For a long time, it was assumed that nuclear explosions are uniquely characterized by high ratios of $^{135}\text{Xe}/^{133}\text{Xe}$ or $^{133\text{m}}\text{Xe}/^{133}\text{Xe}$. However, during a test of noble gas equipment in Freiburg in 2000, for the first time

relatively high $^{135}\text{Xe}/^{133}\text{Xe}$ ratios of nearly 10 were measured in ambient air (AUER *et al.*, 2004). The estimated upper ratios of $^{133\text{m}}\text{Xe}/^{133}\text{Xe}$ were consistent with releases from a nuclear power reactor during the start-up phase (BOWYER *et al.*, 2002) (during the start-up phase the radioisotope ratios are different from the equilibrium ratios). This indicated that single isotopic ratios are not sufficient for discrimination of nuclear tests from civil sources and also that the station specific history of concentrations and atmospheric backtracking must be taken into account. In order to classify a measurement as related to a nuclear explosion, detection of more than two isotopes provides the most reliable evidence (KALINOWSKI *et al.*, 2010). This also sets very specific and stringent requirements to the detection capabilities of the equipment.

During the design phase of the IMS, a set of minimum requirements for IMS radioxenon systems has been defined by the policy-making organ (Preparatory Commission) of the CTBTO (see Table 1). These requirements partly focus on operational requirements of the IMS network, but also specify the detection requirements for IMS radionuclide stations,

Table 1

Minimum requirements for IMS radioxenon systems

Characteristics	Minimum requirements
Air flow	0.4 m ³ /h
Total volume of sample	10 m ³
Collection time	≤24 h
Measurement time	≤24 h
Time before reporting	≤48 h
Reporting frequency	Daily
Isotopes measured	$^{131\text{m}}\text{Xe}$, $^{133\text{m}}\text{Xe}$, ^{133}Xe , ^{135}Xe
Measurement mode	Beta–gamma coincidence or high-resolution gamma spectrometry
Minimum detectable concentration ^a	1 mBq/m ³ for ^{133}Xe
State of health	Status data transmitted to International Data Centre
Communication	Two-way
Data availability	At least 95%
Down-time	No more than 7 consecutive days, No more than 15 days annually

^a Minimum detectable concentrations for the other isotopes are not defined here since they critically depend on the detection system used

Table 2
Radioxenon systems developed for the International Monitoring System

System	Developer
Automatic Radioanalyzer for Isotopic Xenon (ARIX)	Khlopin Radium Institute (KRI), Russian Federation
Automated Radioxenon Sampler-Analyzer (ARSA)	Pacific National Northwest Laboratories (PNNL), USA
Swedish Automatic Unit for Noble Gas Acquisition (SAUNA)	Totalförsvarets Forskningsinstitut (FOI), Sweden
Système de Prélèvement Automatique en Ligne avec l'Analyse du Xénon (SPALAX)	Departement Analyse, Surveillance, Environnement du Commissariat à l'énergie atomique (CEA/DASE), France

which basically condense to the requirement of the aim of a 90% detection probability of a nuclear explosion of 1 kT TNT equivalent within 14 days after the explosion (SCHULZE *et al.*, 2000). The specific needs of IMS radioxenon stations set out in the minimum requirements triggered development of new types of equipment since the mid-1990s. Within only a few years, four new radioxenon systems have been developed, which all were explicitly designed to fulfil the requirement of IMS stations (Table 2). In addition to the development of automated and stationary equipment, mobile sampling and measurement units have been developed. This type of equipment was specifically developed for on-site inspections and is based on similar principles of the IMS-type equipment (CTBT/PTS/TR/2007-1). Furthermore, the development of IMS equipment also triggered development of new laboratory equipment.

3. Technical Principles of Noble Gas Measurement

3.1. Sampling and Processing of Radioxenon

The sampling principle of all IMS noble gas systems is extraction of xenon from atmospheric air by gas separation, purification and concentration on various adsorption and gas separation media. The sample consists mainly of stable xenon which is an atmospheric trace constituent with a concentration of 0.087 ppm by volume (ppmv). The commonly used main mechanism to extract xenon from air is adsorption on activated charcoal: xenon is more strongly absorbed on activated charcoal than the major atmospheric gas constituents. By passing air through a column filled with activated charcoal and by adjusting temperature and flow rate, xenon can be completely

separated from the air. Adsorption is stronger at low temperatures, which is why usually laboratory systems but also the ARSA and ARIX systems perform the adsorption at low temperatures, typically below -100°C . Cooling is however not a necessary condition for xenon adsorption. The relatively large amount of charcoal in the SAUNA sampling system allows sampling also with moderate cooling to -5°C , mainly for moisture removal, using thermoelectric elements (RINGBOM *et al.*, 2003). In the SPALAX system, the air is pre-enriched in Xe with a semi-permeable membrane to 1 ppmv relative to the atmospheric level of 0.087 ppmv (FONTAINE *et al.*, 2004). Due to this pre-enrichment, relatively large amounts of xenon (corresponding to 80 m³ of air for a 24-h sampling period) can be sampled without cooling of the charcoal. In addition, in both systems sampling is switched between two parallel charcoal columns (6-h cycles for SAUNA and 2-h cycles for SPALAX) in order to compensate the less effective adsorption at high temperatures and to avoid a breakthrough of xenon. These subsamples are combined, in order to obtain composite samples with sampling times of 12 h (SAUNA) and 24 h (SPALAX).

After sampling, in order to remove the adsorbed xenon from the activated charcoal trap, the traps are heated typically to about 250–300°C and flushed with an inert purging gas like He or N₂. The purging gas acts as carrier of the collected xenon gas for further processing and measurement. For continuous sampling, two sampling lines are operated in parallel. After this first concentration step, the sample needs further treatment in order to remove trace impurities like Rn, H₂O and CO₂ and to increase the xenon concentration in the sample. This is done by different system specific arrangements of further gas traps.

3.2. Measurement of Radioxenon Activity Concentrations

Activity measurement is either done by high-resolution gamma-spectrometry or beta-gamma coincidence spectrometry. Currently, only the SPALAX system uses a high-resolution (HPGe) gamma spectrometry system (FONTAINE *et al.*, 2004), the other systems all use the beta-gamma coincidence method.

In high resolution gamma spectrometry systems, the isotopes are quantified by their major gamma lines (Table 3). Due to the high selectivity of the sampling process only radioxenon and small remnants of atmospheric radon enter the detection system (more details on radon contamination below). Therefore, the resulting spectrum has relatively little complexity and peak location and identification is simplified by the limited number of possible peaks. However, the relatively low intensities of the gamma emissions for the decays of $^{131\text{m}}\text{Xe}$ and $^{133\text{m}}\text{Xe}$ of 1.95% and 10.0%, respectively, make the detection of these nuclides less sensitive than the detection of ^{135}Xe and ^{133}Xe . Analysis of X-rays helps to increase the sensitivity also for the meta-stable isotopes, however since $^{131\text{m}}\text{Xe}$ and $^{133\text{m}}\text{Xe}$ X-rays have the same energy, X-ray analysis alone gives only an integral measurement of $^{131\text{m}}\text{Xe}$ and $^{133\text{m}}\text{Xe}$. Therefore, distinction between the two nuclides has to be made based on the gamma activity or detection limits based on gamma activity measurement. Here in particular the higher intensity of $^{133\text{m}}\text{Xe}$ gamma emissions can be used for the quantification of $^{131\text{m}}\text{Xe}$, using the information on the absence or presence of $^{133\text{m}}\text{Xe}$. The average minimum detectable activities (MDA), for example reached with the detector of the SPALAX system installed at the IMS radionuclide station at Schauinsland for a 22-h 40-min. measurement, are 600 mBq for $^{131\text{m}}\text{Xe}$, 10 mBq for ^{133}Xe , 132 mBq for $^{133\text{m}}\text{Xe}$ and 47 mBq for ^{135}Xe if only gamma peaks are used for analysis. If only X-rays are present in the spectrum, probabilities for the occurrence of $^{131\text{m}}\text{Xe}$ and $^{133\text{m}}\text{Xe}$ can be given, based on a Bayesian statistical analysis (ZÄHRINGER and KIRCHNER, 2008).

For beta-gamma coincidence systems, the coincident decays listed in Table 3 are utilized for measurement. The noble gas systems currently deployed at IMS stations use a combination of a

plastic scintillator tube as beta detector, which is also used as sample container, which is surrounded by a NaI(Tl) gamma detector. The coincidence measurement leads to a significant reduction of background; e.g., background count rates for an 18-h measurement are typically as low as 0.001 counts per second in the energy interval 70 to 90 keV, depending on the ambient background of the detector location.

Two different types of beta-gamma techniques have been used in IMS systems thus far. The “beta-gamma-energy correlation” method measures the energies of gamma rays and X-rays as well as the energies of beta-particles and conversion electrons. This is of particular relevance for the distinction of $^{131\text{m}}\text{Xe}$ and $^{133\text{m}}\text{Xe}$ X-rays, since by energy analysis, the conversion electrons of the $^{131\text{m}}\text{Xe}$ and $^{133\text{m}}\text{Xe}$ decay can be measured, which have relatively high intensities of 56% and 58%, respectively (REEDER and BOWYER, 1998). Thus the detection limit for these isotopes is strongly improved. This method first used in the ARSA system is also deployed in the SAUNA system (RINGBOM *et al.*, 2003) and will be used in the newest generation of the ARIX system. The typical MDA reached with this method for an 18-h measurement is below 10 mBq for all four xenon isotopes. This neglects, however, the considerable memory effect of these systems (see below), which can increase the MDA significantly after a high activity measurement.

In the so-called “beta-gated gamma systems,” as used in the earlier versions of the ARIX system, the detector only records gamma energy pulse height spectra but no beta energy spectra. The beta or conversion electrons pulses are only used to trigger recording of the gamma or X-rays. For this reason, $^{131\text{m}}\text{Xe}$ and $^{133\text{m}}\text{Xe}$ cannot be separated by energy analysis. Instead the different half-lives are used to analytically separate the two nuclides. The analytical power of this method is, however, strongly limited due to the fact that a long measurement time is required to separate $^{131\text{m}}\text{Xe}$ and $^{133\text{m}}\text{Xe}$ (half-lives 11.84 days and 2.19 days). For this reason, the beta-gated gamma method has recently been abandoned for the ARIX system, which in its newest version also uses a detector-based on the beta-gamma-energy correlation method.

Table 3
Characteristic energies for the decay of ^{131m}Xe , ^{133}Xe , ^{133m}Xe and ^{135}Xe [ENSDF]

	Decay energy (keV)	Branching ratio (%)
^{131m}Xe		
X-rays	29.46	15.4
	29.78	28.6
	33.60	10.2
	34.61	1.85
Gamma rays	163.93	1.95
Conversion electrons	129.4	61
Coincident decays	Sum of 29.46 to 34.61 keV X-rays and 129 keV e^-	56.1
^{133}Xe		
X-rays	30.62	14.1
	30.97	26.2
	35.00	9.4
	36.01	1.7
Gamma rays	80.99	37.0
Conversion electrons	45	55.1
Betas (maximum energy)	346	100
Coincident decays	31.63 keV X-ray + 45 keV e^- + 346 keV beta	48.9
	80.98 keV gamma + 346 beta	37.2
^{133m}Xe		
X-rays	29.46	16.1
	29.78	29.8
	33.60	10.6
	34.61	1.9
Gamma rays	233.2	10.0
Conversion electrons	198.7	64
Coincident decays	Sum of 29.46–34.61 keV X-rays and 199 keV e^-	58.4
^{135}Xe		
X-rays	30.62	1.45
	30.97	2.69
	35.00	0.97
	36.01	0.185
Gamma rays	249.8	90
	608.2	2.90
Conversion Electrons	214	5.7
Betas (maximum energy)	910	100
Coincident Decays	31.63 keV X-ray + 214 keV e^- + 910 keV beta	5.7
	249.8 gamma + 910 keV beta	90
^{214}Pb		
X-rays	74.82	4.8
	77.11	8.0
	86.83	1.0
	87.35	1.8
	89.78	0.67
	90.07	0.17
Gamma rays	241.95	7.43
	351.95	37.6
^{214}Bi		
X-rays	79.29	0.98

Also shown are gamma and X-ray energies from the ^{222}Rn daughter ^{214}Pb and ^{214}Bi , which may cause interference with radionuclide detection

Common to all beta–gamma systems is the relatively low resolution of the measurement, both in the beta/conversion electron detection (resolution requirements for IMS systems: better than 40 keV at

129 keV energy) and in the gamma detection (IMS resolution requirement: better than 15% FWHM at 80 keV) (CTBT-PTS-INF.921-Rev.3). These detectors can have a memory effect of around 5%, since

the porosity of the plastic scintillator material allows xenon to diffuse through the walls of the detector (BEAN *et al.*, 2007). In order to avoid false detection of radion xenon, in particular for a sample following a high activity measurement, the memory effect has to be accounted for by performing a background measurement prior to each sample measurement. The background counts from the background measurement must be subtracted from the counts determined in the sample measurement. This however, leads to a degradation of the minimum detectable activity in case of a measurement following a high activity sample. The high resolution gamma detector of the SPALAX systems does not have such a memory effect, since the sample container used for the activity measurement consists of aluminium.

For all types of detection systems, daughter activities of ^{222}Rn in the sample can cause interference with radion xenon signals. Ambient levels of ^{222}Rn in ground-level air are typically in the order of 10 Bq/m^3 (UNSCEAR, 2000). The half-life of ^{222}Rn is long enough (3.82 days) to survive the several-hour sample processing and thus it can enter the detector cell if not separated from the sample. The half-lives of the ^{222}Rn daughters ^{214}Bi ($T_{1/2} = 19.9\text{ min}$) and ^{214}Pb ($T_{1/2} = 26.8\text{ min}$), which have gamma and X-ray energies interfering with the radion xenon detection (Table 3), are short enough compared with the counting times of several hours to be in equilibrium with the radon, therefore any contamination with radon can cause a significant increase in gamma background. Due to its high atmospheric concentration, separation of ^{222}Rn has to be efficient, in all systems, the concentration in a sample is typically reduced by a factor of 10^5 .

For determination of the activity concentration, the air volume of the sample has to be determined. This is done by quantification of the stable xenon volume in the activity counting cell. Since the concentration of stable xenon in the atmosphere is constant, the volume of stable xenon (V_{Xe}) can be converted into the corresponding air volume (V_{air}) by:

$$V_{\text{air}}[\text{m}^3] = V_{\text{Xe}}[\text{cm}^3]/0.087.$$

The stable xenon volume measurement is done either with a gas chromatograph or with a Thermal

Conductivity Detector (TCD). The uncertainty of the stable gas measurement is typically 10–15%.

The activity concentration C in Bq/m^3 is calculated by:

$$C = \frac{\text{Sample activity [Bq]}}{\text{Xenon volume [cm}^3]} \times 0.087 \frac{[\text{cm}^3]}{[\text{m}^3]}.$$

4. Operational Experience during the Last 10 Years

4.1. The IMS Noble Gas Equipment Test

In parallel to the development of equipment, the Provisional Technical Secretariat (PTS) of the Preparatory Commission (PrepCom) for the CTBTO started the Noble Gas Equipment Test in 1999 (CTBT/PTS/INF.162), which was designed to ensure compliance of the newly developed equipment with the analytical and operational requirements of the IMS. In the course of the test, this programme was then extended to the International Noble Gas Experiment (INGE), which also includes aspects of data analysis, data interpretation, certification and operation of the radion xenon component of the IMS. The noble gas equipment test, should, aside from a technical assessment of the state of the art of noble gas technology, facilitate the transition of operation of the systems in a laboratory environment to automated routine operation in a remote environment under typical IMS-type operational conditions.

The Noble Gas Equipment Test has been divided into an equipment development phase (Phase I), an assessment of the analytical performance of the systems (Phase II) and a test of the operational (and also analytical) performance of the systems and of the network (Phase III), the latter phase being a still ongoing activity. The basic aims of the test show that:

- Noble gas monitoring is a reliable technique for monitoring nuclear tests (Phase II).
- The minimum requirements (Table 1) can be fulfilled (Phase II).
- The systems are suitable for operation under typical IMS conditions (Phase III).

The actual testing of the systems started with Phase II, during which the systems have been installed and operated side-by-side at the German Federal

Office for Radiation Protection (BfS) in Freiburg. Phase II started with the installation of an ARSA system in October 1999 and lasted until February 2001. The systems were tested in routine operation, measuring the ambient radionuclide concentrations, but also by performing spike experiments, i.e., injection of radioactive xenon samples containing mixtures of $^{131m}\text{Xe}/^{133}\text{Xe}$ and $^{135}\text{Xe}/^{133}\text{Xe}$ either into the air sampling systems or directly into the detectors of the systems. Occasionally, archived samples have also been re-analyzed at the noble gas laboratory of the BfS. The systems showed a good overall agreement within each other and with the measurements performed at the BfS laboratory. The experiment showed that all systems measured the activity of radioactive xenon isotopes with an accuracy approaching 10–20%. The minimum detectable activity concentrations for the isotope ^{133}Xe ranged between 0.1–1 mBq/m³, showing that all four test systems fulfil the sensitivity requirements of the IMS. The measurement of archived samples with the BfS laboratory system also demonstrated the feasibility of independent re-analysis with a laboratory-based system for quality control. The test phase also provided a wealth of high resolution data in an area with a high density of nuclear facilities. New findings during this phase were the observation of xenon peak concentrations to over 100 mBq/m³ with a duration of 12 h and less and the observation of relatively high $^{135}\text{Xe}/^{133}\text{Xe}$ ratios of up to 10. A detailed description and summary of the procedures and results of this phase of the test are given in AUER *et al.* (2004).

4.2. Field Testing and First Network Operation Experience

The transition of Phase II to Phase III of the test was accompanied by a transition of prototype systems provided by the system developers to commercial “off-the-shelf” type systems. Regarding the SAUNA and SPALAX systems, these new systems were provided by the commercial companies Gammadata Instrument AB (<http://www.gammadata.se>) for SAUNA and Environnement S.A. (<http://www.environnement-sa.com/>) for SPALAX, the new ARIX is still provided by its developer (Khlopin Radium Institute, <http://www.khlopin.ru>). The ARSA system is not provided as a

commercial system, however major technological principles of this system, such as the beta–gamma coincidence detection method, have also been integrated in other systems.

Phase III of the Noble Gas Equipment Test is related to the testing of the operational performance of the systems installed at IMS radionuclide stations, i.e., examination of the verification of the operational parameters in the field, as well as the assessment of operational reliability of the systems. Besides this, the Phase III exercise comprises network-related aspects of the noble gas experiment such as establishment of a network-wide maintenance strategy, preparation of operational manuals, development of certification requirements, establishment of a QA/QC system, atmospheric transport modelling, as well as the development of a categorization scheme for noble gas monitoring. A further aspect of Phase III was the set-up of “mini-networks” in North America, Europe and Asia with four systems per mini-network in order to assess correlations between stations in areas with a relatively high density of nuclear facilities.

With regard to equipment testing, Phase III was divided into three sub-phases to reflect different stages of the field experience: Phase III/a consisted of the installation and tuning of the systems in the field. During this stage the system providers had unrestricted access to their systems for tuning and troubleshooting after installation. Next, Phase III/b was the first period where systems were operated by the station operator and the PTS. Per system type, a minimum time of 6 months for Phase III/b operation was considered necessary in order to collect sufficient data for the evaluation. After each system type has successfully completed Phase III/b, more systems have been installed and are running under Phase III/c with the scope of further testing and evaluation of equipment and the other Phase III/c tasks mentioned above.

The systems operating in Phase III (currently operated systems are shown in Fig. 1) show improved operational parameters compared to the Phase II prototype systems (Table 4).

Furthermore, IMS minimum requirements related to data transmission and operational performance are being assessed during Phase III: All data relevant for monitoring the operational status are transmitted and systems allow for two-way communication between

Table 4
Operational parameters of systems which were participating in Phase III testing

Characteristics	Minimum requirements	SPALAX	SAUNA	ARIX
Airflow	0.4 m ³ /h	15–20 m ³ /h	1.2 m ³ /h	1.6 m ³ /h
Total volume of sample	10 m ³	50–75 m ³	25–30 m ³	36 m ³
Collection time	≤24 h	≤24 h	12 h	12 h
Measurement time	≤24 h	≤24 h	11 h 10 min	18 h
Time before reporting	≤48 h	≤48 h	30 h	34 h
Reporting frequency	Daily	Daily	2 samples per day	2 samples per day
Isotopes measured	^{131m} Xe, ¹³³ Xe, ^{133m} Xe, ¹³⁵ Xe	^{131m} Xe, ¹³³ Xe, ^{133m} Xe, ¹³⁵ Xe	^{131m} Xe, ¹³³ Xe, ^{133m} Xe, ¹³⁵ Xe	^{131m} Xe, ¹³³ Xe, ^{133m} Xe, ¹³⁵ Xe
Measurement mode	β - γ -coincidence or high-res- γ - spectrometry	High-res- γ - spectrometry	β - γ -coincidence	β - γ -coincidence
MDC for ¹³³ Xe	1 mBq/m ³	0.2–0.6 mBq/m ³	0.2–0.4 mBq/m ³	0.2–0.3 mBq/m ³

All values except MDCs are nominal operational values given by system suppliers and were verified during Phase III operation. MDC ranges were assessed during testing

station operators and the PTS. Data availability, down time and related reasons for operational failures are assessed. Data availability has been defined as the number of spectra that are received in a considered period and contain the essential information for spectral processing, compared to the total number of spectra expected for the considered period. Down-time causes are categorized according to the component where a failure occurred, as there are: (1) sampling and gas processing system, (2) nuclear measurement system, (3) gas quantification system, (4) power system, (5) computer and software, (6) human errors, (7) organisational problems and (8) other.

Assessment during the early years of Phase III operation has shown that the main causes of down-time are related to problems in the HPGe detector cooling of the nuclear measurement system, in the gas processing system of the equipment and in the software. Whereas failures in the detector resulted in down-time periods of several weeks to months, software problems and failures in the gas-processing system could be fixed within a couple of days by the station operator, provided that appropriate spare parts were present at the station. The PTS together with the system suppliers are establishing a network-wide maintenance strategy within the scope of Phase III/c to overcome such deficiencies. With a view to data availability during Phase III the systems have shown that they are able to operate according to IMS requirements over several months.

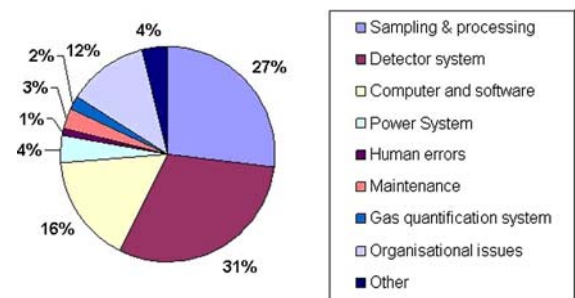


Figure 2
Network wide down-time causes, categorized according to the sub-systems where they occurred

However, since the maintenance and data analysis system of the IMS noble gas network is not yet fully operational, quick and effective repair mechanisms are not fully in place and down-times can be several months. For the total IMS noble gas network, down time was 2,487.5 days out of total 9,605 days or 25.9% of operational days between July 2004–July 2008. An overview of typical error causes and their frequencies is shown in Fig. 2.

5. Operational Experience and Data from the Noble Gas Station at the IMS Radionuclide Site at Schauinsland

The IMS location RN33 at Schauinsland, Germany (47°54'N, 7°54'E) is one of the forty radionuclide stations which host, beside a particulate

system, noble gas equipment. The site is located on a mountain ridge (approx. 1,200 m asl) in the Black Forest overlooking the Rhine Valley. It has a long history in monitoring of nuclear explosions. Initially used for measurement of cosmic radiation in the 1950s, already in 1953 debris from atmospheric nuclear tests was detected (SITTKUS, 1955) and afterwards a programme to continuously monitor radioactivity from natural sources and fission products from weapons test fall-out has been established which continues to date (STOCKBURGER and SITTKUS, 1966; BIERINGER and SCHLOSSER, 2004). Also at the site an automated IMS particulate station (RASA) is operated which was certified by the PTS in 2004 (ZÄHRINGER *et al.*, 2008). Radioxenon measurements have been continuously performed by the BfS at the Schauinsland since 1980 using manually operated equipment. The sampling equipment consists of absorbers filled with activated charcoal and cooled with liquid nitrogen. Sampling time is 1 week, the analysis of the samples, which contain xenon from approximate 10 m³ air is done at the central BfS laboratory in Freiburg. The minimum detectable ¹³³Xe concentrations for these samples are approximately 1 mBq/m³. The median ¹³³Xe activity concentration in ground level air at this site over the last 28 years is 4.6 mBq/m³, with peak concentrations reaching approximately 40 Bq/m³ observed after the Chernobyl accident (Fig. 3).

In February 2004 a SPALAX system was installed at Schauinsland (Fig. 4). Since then, the system has been operated almost continuously (Fig. 5) with a major interruption of operation from 15.04.2006 until 24.02.2007 due to a breakdown of the electrical detector cooling system. Another long interruption occurred in spring 2008 due to a compressor failure and the delivery time for spare parts.

The ¹³³Xe concentrations measured from 2004 to 2008 are shown in Fig. 6. These data have been analyzed with the Aatami nuclear spectral analysis software (CTBTO, Preparatory Commission, 2007). Also shown in the figure are the weekly ¹³³Xe concentrations measured with the laboratory system of the BfS during the corresponding period. The ¹³³Xe activity concentrations reported by the BfS system are integral values of the activity of all radioxenon isotopes. Since the atmospheric radioxenon

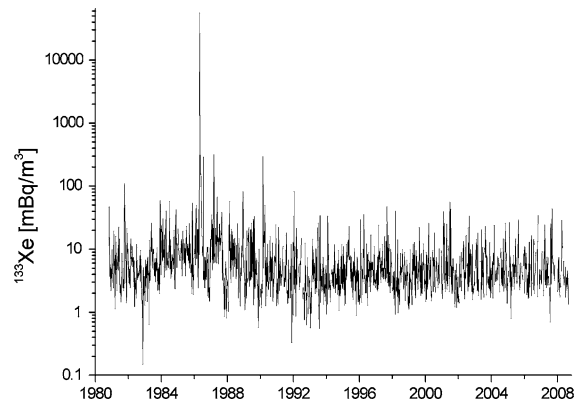


Figure 3
¹³³Xe concentrations measured at Schauinsland, Germany, since 1980. The large peak in 1986 is due to xenon released during the Chernobyl accident



Figure 4
SPALAX noble gas system installed at the Radionuclide Station RN33 at Schauinsland, Germany

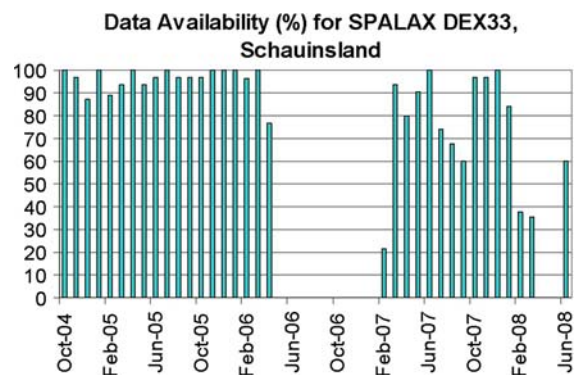


Figure 5
Data availability of SPALAX at the Radionuclide Station RN33 Schauinsland, Germany since start of operation

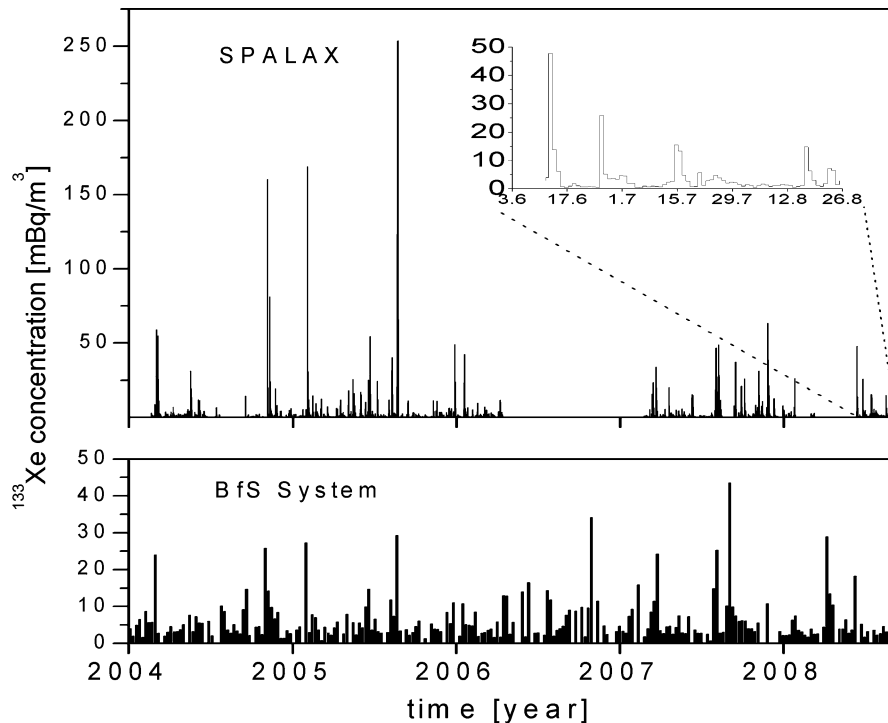


Figure 6

^{133}Xe activity concentration time series measured at Schauinsland with SPALAX and the BfS noble gas system. The *insert* shows an enlarged time interval for the period 03.06.2008 to 26.08.2008

concentrations are strongly dominated by ^{133}Xe , the error due to neglecting ^{135}Xe , $^{133\text{m}}\text{Xe}$ or $^{131\text{m}}\text{Xe}$ is small. This is supported by the SPALAX data which indicate that the sum of the activity of $^{131\text{m}}\text{Xe}$, $^{133\text{m}}\text{Xe}$ and ^{135}Xe in all samples is less than 10% of the activity of ^{133}Xe , showing that taking the activities measured with the BfS system as ^{133}Xe activity is a sufficiently good approximation. As may be expected, the longer sampling duration of the BfS systems, combined with the short duration of ^{133}Xe peaks, results in lower peak concentrations of the BfS system relative to the concentrations measured by SPALAX.

For comparison of the SPALAX results with the BfS data, the SPALAX data have been combined to weekly values taking into account the radioactive decay: For each SPALAX sample, the activity concentration has been decay-corrected to the end of the BfS sampling cycle and the mean of the seven daily samples has then been used to calculate the mean activity during the corresponding 7-day period, assuming a constant concentration during that period.

For the comparison only data concentrations above 2 mBq/m^3 (i.e., approximate 2 times higher than the MDC of the BfS detection system) have been taken into account. The two data sets are clearly correlated (Fig. 7), although there is a bias of 0.73 ± 0.08 of the concentrations measured by the BfS system relative to the SPALAX system. The bias was estimated with a weighted least-squares fit method which takes into account uncertainties in both coordinates (REED, 1988). Note that the fit yields, within the uncertainties, the same results if the two highest data points are not allowed for.

Thirty-seven archived SPALAX samples have also been re-analyzed at the noble gas laboratory of the BfS (Fig. 8). A similar bias between SPALAX measurements and BfS system, which already has been observed in the comparison of independent samples measurements, is also observed in the re-analysis of the archive bottles. Here, the bias is 0.81 ± 0.01 . The bias is most likely caused by systematic differences in the determination of the volume of stable xenon in the sample between

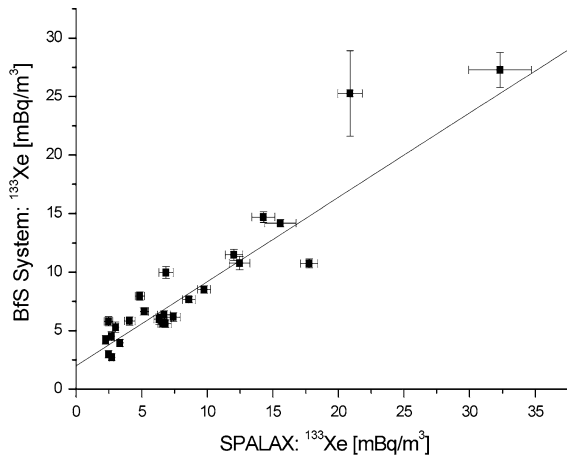


Figure 7

^{133}Xe weekly activity concentrations measured by the SPALAX system and by the BFS laboratory system. The line shows the uncertainty weighted least-squares fit of the data. The concentrations from SPALAX are calculated from daily values

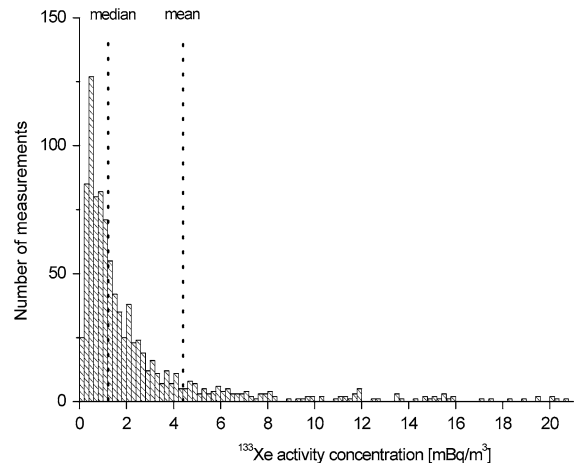


Figure 9

Frequency distribution for atmospheric ^{133}Xe concentrations measured with the SPALAX system at Schauinsland. Dotted lines indicate the median and the mean of the data set

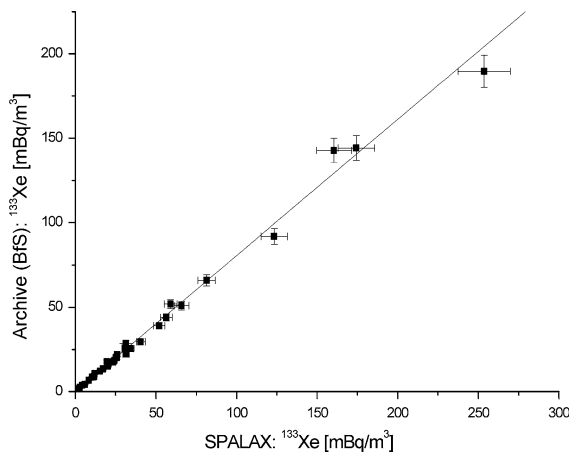


Figure 8

^{133}Xe concentrations measured by SPALAX and compared with re-measurements of the archived samples with the BFS laboratory system. The line shows the uncertainty weighted linear least-squares fit of the data

SPALAX and the BFS laboratory, since the re-analysis of the stable xenon concentration frequently yielded a higher stable xenon content than the analysis by SPALAX. The similar bias of the two comparisons shows on the other hand the good reproducibility of the atmospheric radionon measurements by independent samplers. Taking into account the bias between the two measurement methods, the data from the BFS long term time series can be used to characterize the radionon

background for evaluation of the data measured with the SPALAX system.

The concentrations measured by the SPALAX system range from below 0.1 mBq/m^3 in only three of the 995 samples of the data series up to $(253 \pm 16) \text{ mBq/m}^3$. It is worth noting that in only two of the samples, the activity was below the critical limit for detection, showing that the sensitivity of the SPALAX system is well suited for measurement of even the lowest ambient levels of radionon for a location such as Schauinsland. The range and distribution of concentrations are shown in Fig. 9. The typical background level for ^{133}Xe ranges between 0.2 and 1.3 mBq/m^3 . The median is at 1.28 mBq/m^3 , the mean at 4.4 mBq/m^3 . The peaks of ^{133}Xe activity concentrations have a typical duration of one to three days as shown in the insert of Fig. 6.

In 10 of the total of 995 samples, the activity of all four isotopes was above the critical limit for detection. Those multiple isotope detections were associated with relatively high ^{133}Xe concentrations ranging between 6 mBq/m^3 and 175 mBq/m^3 and a median concentration of 41 mBq/m^3 with a mean uncertainty of 10%. The $^{133\text{m}}\text{Xe}/^{133}\text{Xe}$ ratios range between 0.015 and 0.13 (mean of the uncertainty 23%), the $^{135}\text{Xe}/^{133}\text{Xe}$ ratios range between 0.007 and 0.069 (mean of the uncertainty 38%) and the $^{133\text{m}}\text{Xe}/^{131\text{m}}\text{Xe}$ ratios range between 0.7 and 3.3 (mean of the uncertainty 37%). Those ratios are well

within the expected values from reactor operations or with emissions from radiopharmaceutical production plants (KALINOWSKI *et al.*, 2010).

6. Conclusion and Outlook

The installation of the IMS for the CTBTO triggered new developments of equipment for measurement of radionoble isotopes during the last decade. Compared to the equipment which was previously available and which was based on manually operated laboratory systems, significant progress has been made. The equipment which has been developed is automated and measures with a high sensitivity the four isotopes ^{131m}Xe , ^{133}Xe , ^{133m}Xe and ^{135}Xe . The analytical capabilities of these newly developed systems fulfil and even exceed the stringent detection requirements of the IMS. From this point of view, these systems have proven to be excellent tools for the verification system of the CTBT. A major requirement for monitoring the CTBT is also operational stability and reliability, in particular since the systems are frequently operated at remote sites with limited infrastructure. The operational characteristics of the systems under “field conditions” at various locations worldwide have been tested during the last 5 year and provided insight also into the technical long-term performance.

As of October 2008, 16 radionoble systems are sending data to the PTS (7 SPALAX systems, 7 SAUNA systems and 2 ARIX), additional six stations are under installation, hence about 50% of the IMS noble gas network will be operational soon. Overall, the performance and reliability of the systems operation is high, however the evaluation of system down-times is difficult at present, since the structures and logistic measures for maintenance of these systems are not fully operational yet. Analysis in particular of reoccurring failures, may allow even further enhancement of the operational reliability.

The data produced by the already installed noble gas systems provide an unprecedented coverage of worldwide radionoble activity concentrations. This allows a comprehensive assessment of global background data in regions with substantially different

distribution and strength of sources. The density of the network also allows for investigation of the correlation of signals measured at neighbouring stations in order to improve the determination of the source location. These data are the basis for a future scheme for the evaluation and analysis of radionoble concentrations with respect to their significance as an indicator for nuclear tests.

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