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Environmental Radioxenon Levels in Europe: a Comprehensive Overview

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Abstract—Activity concentration data from ambient radioxenon measurements in ground level air, which were carried out in Europe in the framework of the International Noble Gas Experiment (INGE) in support of the development and build-up of a radioxenon monitoring network for the Comprehensive Nuclear-Test-Ban Treaty verification regime are presented and discussed. Six measurement stations provided data from 5 years of measurements performed between 2003 and 2008: Longyearbyen (Spitsbergen, Norway), Stockholm (Sweden), Dubna (Russian Federation), Schauinsland Mountain (Germany), Bruyères-le-Châtel and Marseille (both France). The noble gas systems used within the INGE are designed to continuously measure low concentrations of the four radioxenon isotopes which are most relevant for detection of nuclear explosions: ^{131m}Xe, ^{133m}Xe, ^{133}Xe and ^{135}Xe with a time resolution less than or equal to 24 h and a minimum detectable concentration of ^{133}Xe less than 1 mBq/m^3. This European cluster of six stations is particularly interesting because it is highly influenced by a high density of nuclear power reactors and some radiopharmaceutical production facilities. The activity concentrations at the European INGE stations are studied to characterise the influence of civilian releases, to be able to distinguish them from possible nuclear explosions. It was found that the mean activity concentration of the most frequently detected isotope, ^{133}Xe, was 5–20 mBq/m^3 within Central Europe where most nuclear installations are situated (Bruyères-le-Châtel and Schauinsland), 1.4–2.4 mBq/m^3 just outside that region (Stockholm, Dubna and Marseille) and 0.2 mBq/m^3 in the remote polar station of Spitsbergen. No seasonal trends could be observed from the data. Two interesting events have been examined and their source regions have been identified using atmospheric backtracking methods that deploy Lagrangian particle dispersion modelling and inversion techniques. The results are consistent with known releases of a radiopharmaceutical facility.

Key words: Comprehensive Nuclear-Test-Ban Treaty, low-level environmental radioactivity measurements, noble gas, European air, radioxenon, nuclear verification.

1. Introduction

The Comprehensive Nuclear-Test-Ban Treaty (CTBT), which was opened for signature in 1996, is a key element in the non-proliferation of nuclear weapons and a crucial basis for the pursuit of nuclear disarmament as it bans any kind of nuclear explosion.

For its verification several techniques are provided. Beside waveform measurements (seismic, hydro-acoustic and infra sound), radionuclides are measured (radioactive particulates and noble gases) and atmospheric transport models are used to identify the possible source regions of radioactive particulate or noble gas detections.

When the International Monitoring System (IMS) network is completed there will be 321 stations

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worldwide; among them 80 permanently sampling and measuring radioactive particles. At least 40 of these will also be equipped with noble gas measurement systems (UNGA, 1996; DAHLMAN and MYKKELTVEIT, 2009).

Noble gas monitoring is a fundamental and highly sensitive technique for the detection of underground or underwater nuclear explosions. Of all the verification technologies, it is, together with radionuclide particulate monitoring, the only technique that has the potential to provide unmistakable proof of a nuclear explosion (DE GEER, 1996).

To establish this global noble gas network, fully automated radioxenon measurement systems had to be developed, as no commercial systems were available when the Treaty was opened for signature (AUER *et al.*, 2004). Four countries, France, Russia, Sweden and USA, all with experience of atmospheric xenon measurements, offered to develop such systems. With the Provisional Technical Secretariat (PTS) for the CTBT Organisation (CTBTO) and the German Federal Office for Radiation Protection (Bundesamt für Strahlenschutz, BfS), they participate in the International Noble Gas Experiment (INGE) project (AUER *et al.*, 2004).

Under the auspices of the INGE, these systems are now undergoing tests at worldwide locations and send their results to the International Data Centre (IDC) in Vienna for processing and analysis (SAEY and DE GEER, 2005). All four systems that have been developed are able to measure the four radioxenons of interest for CTBT verification: ^{131m}Xe, ^{133m}Xe, ^{133}Xe and ^{135}Xe with the half-lives of 11.9 days, 2.19 days, 5.24 days and 9.14 h, respectively.

The design criterion for all of the equipment is that the minimum detectable concentration (MDC) of ^{133}Xe should be 1 mBq/m^3 or less for a 24 h sampling period. Supporting this, internal documentation of the PTS prescribe an average air flow above 0.4 m^3/h, where the actual sampling rate should never fall outside ±30% of the average during the sampling period. The total air volume sampled should be at least 10 m^3 per day. The 1 mBq/m^3 MDC for ^{133}Xe was state-of-the-art in the mid-1990s when the requirement was set, but has since then improved by almost a factor of ten.

For this study data from three different noble gas systems were used, the Russian ARIX-II (Analyzer of Radioactive Isotopes of Xenon), based on a beta-gated gamma nuclear measurement system (DUBASOV *et al.*, 2005), the French SPALAX (Système de Prélèvement d'air Automatique en Ligne avec l'Analyse des radio-Xénons), which is based on high-resolution gamma spectroscopy (FONTAINE *et al.*, 2004) and the Swedish SAUNA-II (Swedish Automatic Unit for Noble gas Acquisition), which uses beta-gamma coincidence spectrometry (RINGBOM *et al.*, 2003).

2. *Observation of Environmental Radioxenon*

2.1. *European Radioxenon Measurements in the past: Some History*

To put the new, high time resolution measurements of this paper in relation to other radioxenon measurements performed in Europe in the past, we give in the following an overview about older published measurements of atmospheric radioxenon.

2.1.1 *World War II*

The first atmospheric radioxenon measurements date back to the fall of 1944, at the end of the Second World War and only 2 years after the first successful test of a nuclear reactor in December 1942. Douglas A-26 medium bomber airplanes from the American Ninth Air Force flew low over Germany and sampled air to search for radioxenon fingerprints of possible nuclear reactors operated in Germany and a related weapons programme (ZIEGLER and JACOBSON, 1995).

This operation was set up by a special intelligence unit from the Manhattan Engineer District. The scientific idea of collecting environmental noble gases in the air using large charcoal tubes in the bomb bay of the airplane and then measure the xenon-133 back home in the laboratory, was developed by later Nobel Prize laureate Luis Alvarez. He became the first scientist to develop a radiological air sampling method of overhead reconnaissance. No radioxenon was detected in these campaigns as there were no reactors or nuclear weapons in Germany at that time.

2.1.2 *Germany*

In the 1960s, radioxenon measurements were reported from the University of Heidelberg. Their goal was to measure ^{85}Kr and ^{133}Xe released by the ongoing

nuclear weapon tests in the atmosphere. EHHALT *et al.*, published in 1963 results from a campaign where they indeed observed clear signals from Russian atmospheric tests carried out in the autumn of 1961 (EHHALT *et al.*, 1963). SCHÖLCH *et al.* (1966) suggested that a peak of about 10 mBq/m^3 detected in a 10-day air collection sample in early June 1965 might have been due to the second Chinese atmospheric test, which had been performed about 3 weeks earlier (on 14 May 1965, a nuclear explosion with a yield of 35 kt TNT equivalent in 500 m above ground; MIKHAILOV *et al.*, 1999). From a few ^{133}Xe measurements in late 1964 and early 1965 they also reported a general background of about 5 mBq/m^3 presumably from European reactors.

In the early 1970s the BfS started to continuously monitor the atmospheric ^{133}Xe and ^{85}Kr activity concentrations in Freiburg, Germany (STOCKBURGER *et al.*, 1977). At present the atmospheric activity concentration of ^{133}Xe in ground level air is continuously monitored at seven sites in Germany as part of the "Integrated Monitoring and Information System" (IMIS) (WEISS and LEEB, 1993), the German surveillance program for radioactivity in the environment. In addition, samples are taken at other stations around the globe, e.g., at the Meteorological Research Institute (MRI) in Tsukuba, Japan (IGARASHI *et al.*, 2000b; SCHLOSSER *et al.*, 2003).

The pre-enriched samples taken at the sites are sent in a 1 L aluminium vessel to the central noble gas laboratory at BfS in Freiburg for analysis. The collection time during routine operation is seven days. The total volume sampled is around 10 m^3 of air. The procedures for sampling, enrichment and purification of the noble gas fractions are all manual (IGARASHI *et al.*, 2000a). The integral beta activity of the samples is measured in proportional counters using methane as an additional gas component. This integral counting method gives the total activity of all radioxenons but a separation of the components can be done by decay analysis.

Besides ^{133}Xe, the most abundant radioxenon isotopes observed in environmental samples, contributions of ^{131m}Xe and ^{135}Xe could be determined to a few percent of the total beta-activity. The MDC for ^{133}Xe in routine samples is about 1 mBq/m^3. The longest time series available is from the station in Freiburg (Fig. 1).

During recent years the average ^{133}Xe activity concentration measured in weekly samples at German stations is around 6 mBq/m^3 with large variations between 1 and 100 mBq/m^3 (BMU, 2007). The maximum activity concentration of 106 Bq/m^3 was measured in the daily sample of 1 May 1986 taken at BfS in Freiburg. It originated from the Chernobyl reactor accident.[1]

In the years between 1987 and 1995 the atmospheric ^{133}Xe activity concentration decreased by a factor of around 20. This behaviour is consistent with reported noble gas release data from nuclear power plants in Germany and could be explained by improvements of the nuclear fuel rod cladding and reactor containment systems as well as with longer delay times before the release of noble gases in the atmosphere (BIERINGER and SCHLOSSER, 2004).

2.1.3 Sweden

A system for sampling and analysis of small amounts of radioxenon in ambient air was developed around 1980 by the Swedish Defence Research Agency (FOI, formerly FOA). This was a forerunner to the SAUNA system but used at that time charcoal adsorption at −80°C and high-resolution gamma spectroscopy for detection. During the development phase, 2–3 days samples were periodically taken in Stockholm and during 1982 also at a satellite station at Ljungbyhed in southern Sweden (BERNSTRÖM and DE GEER, 1983). Initially the average detection limit was 1.8 mBq/m^3, which was later reduced by a factor of three by chromatographic suppression of ^{222}Rn. The reported average activity concentration of ^{133}Xe was 8.6 mBq/m^3 at the Stockholm station and 22.6 mBq/m^3 at the satellite station. These were quite high values and occasionally extreme values were measured up to around 250 mBq/m^3. The reason for the high values was excessive emission from a boiling water reactor near Gothenburg, some 420 km southwest of Stockholm and 150 km north of Ljungbyhed. Up to 26 fuel elements exhibited cracks and these elements were

[1] For a long time only Germany was known to have measured radioxenon from the Chernobyl accident. Recently, however, ^{133}Xe concentrations around 1.5 Bq/m^3 were reported from late April 1986 in Cherepovetz, Russia (PAKHOMOV and DUBASOV, 2008).

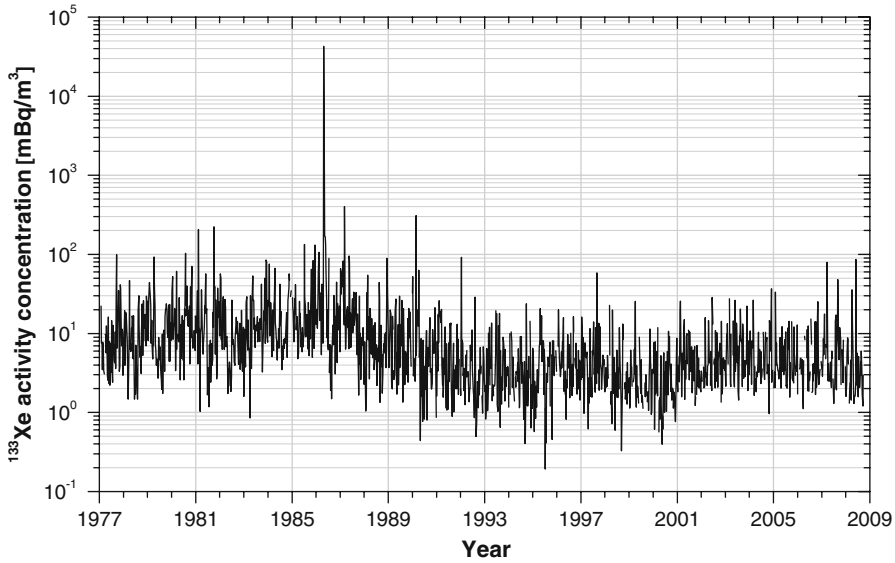

Figure 1
Weekly ^{133}Xe activity concentration of ^{133}Xe in ground level air at Freiburg, Germany

exchanged during the 1981 summer revision. In 1982 the levels then went down to typically below the detection criterion. At the end of October 1980 roundly 100 mBq/m^3 were observed in Stockholm. At that time there were no excessive emissions from Swedish reactors and the signal was interpreted to be due to the last atmospheric nuclear test conducted 2 weeks earlier on 16 October 1980 at Lop Nor, China, with a yield between 150 to 1,500 kt (MIKHAILOV *et al.*, 1999).

The Swedish xenon system was mothballed between 1983 and 1989 but from October 1990 until about 1998 there was a series of measurements from Stockholm and for part of that time also from two satellite systems, one at Ljungbyhed in the south and one in Kiruna in the north. At the satellites pre-processed gas samples were collected and then sent by post in a gas-proof plastic bag to the laboratory in Stockholm for analysis. In October 1990 a 24 mBq/m^3 peak in the xenon activity concentration was observed in Stockholm that, based on careful meteorological backtracking analyses, was believed to be due to a small leak from the last Soviet nuclear test on 24 October 1990 (an eight device shot of in total 40 to 380 kt at Novaya Zemlya, about 2,100 km NE of Stockholm) (MIKHAILOV *et al.*, 1999).

During the period a slowly decreasing trend was observed with an average of 3.0 mBq/m^3 in Stockholm at 59°N, some 0.7 mBq/m^3 in Kiruna at 68°N and some 0.8 mBq/m^3 in Ljungbyhed at 56°N. In late March 1992, a peak at 24 mBq/m^3 was observed in Stockholm. This large activity concentration originated from an accident/incident at the Sosnovy Bor nuclear power plant 100 km west of St. Petersburg and 600 km east of Stockholm. With time the more routine operations of the Swedish xenon stations were phasing out as work increasingly was concentrated on developing the SAUNA system. In preparation for Phase 2 of INGE, when all four systems were to be run in parallel in Freiburg, Germany (see Sect. 2.5), a surveillance period in Stockholm in September 2000 exhibited a mean activity concentration of ^{133}Xe of about 1 mBq/m^3 RINGBOM *et al.* (2003).

2.1.4 *The IMS Noble Gas Network*

There are many significant improvements of the new systems set-up for CTBT verification as compared to the older ones just described:

- Beside the long time series of the BfS, measurements in the past were often performed during short, well-defined time periods. The data presented in this paper are collected over several years and the measurements are ongoing;

- The sampling time during these campaigns was often of the order of several days and upto 1 week or even more. In the INGE network, the collection time resolution is between 8 and 24 h;
- In most of the older measurements, the method was optimized for the detection of ^{133}Xe. Due to shorter cycle times, higher processing capabilities and improved counting technologies, the new INGE systems can detect four different radioxenon isotopes simultaneously with a high sensitivity;
- These systems have a very strong detection capability, with ^{133}Xe MDC to 0.1 mBq/m^3 for 12-h samples;

All these improvements make it possible to perform more accurate isotope ratio analysis for source discrimination, better source identification and meteorological geo-location analyses.

2.2. *Current Set-up of the Measurement Stations in Europe*

For this paper, we used continuous radioxenon data with a high time resolution from six different European locations.

At present, four systems of the INGE network are installed at European IMS stations (Spitsbergen in the Norwegian archipelago Svalbard; Stockholm, Sweden; Dubna, Russia and on the Schauinsland Mountain, near Freiburg, Germany). In Bruyères-le-Châtel, 30 km south of Paris, a system is operated by the French Atomic Energy Commission (Commissariat à l'Énergie Atomique, CEA). The measurements at the Marseille site were performed by Environment S.A (the manufacturer of the French SPALAX system) with two IMS systems purchased by Health Canada, which collected data prior to shipment to their end destinations in Canada (IMS stations in Yellowknife and St. John's). The geographical and sampling system specifications of the six different sites are listed in Table 1.

The Swedish and Norwegian stations are equipped with SAUNA-II systems, the French and German with SPALAX devices and the Russian with ARIX-II equipment.

The SAUNA-II system samples air in 12-h cycles. Then the collected xenon fraction is purified and concentrated during some 7 h before it is counted with the (plastic and NaI) beta-gamma coincidence detector for approximately 11 h. These spectra have been analysed with PTS developed software, based on a net-count method.

The SPALAX system continuously samples air for 24 h per cycle. At the end of such a collection cycle the final purification and transfer into the counting system needs about one more hour before the start of counting on a broad energy high-purity germanium gamma-ray detector for around 23 h. The spectra from Schauinsland were analysed and reviewed with the PTS developed radionuclide analysis and evaluation software AATAMI (2003), those from Bruyères-le-Châtel and Marseille with Canberra's software Genie 2000. The xenon isotopes analysed with Genie 2000 are based on the gamma ray peak information only—the Aatami software also uses additional information from the xenon and caesium X-rays in the 30 keV region.

The ARIX-II system collects air in 12-h cycles. Then the air is purified and concentrated during approximately 4 h before it is counted with the (plastic and NaI) beta-gated gamma detector for

Table 1

Location of the radioxenon measurement systems considered in this study, listed from north to south

Station	Host country	Latitude	Longitude	Altitude (m)	System
Spitsbergen	Norway	78.1°N	15.2°E	220	SAUNA-II
Stockholm	Sweden	59.4°N	18.0°E	40	SAUNA-II
Dubna	Russian Federation	56.4°N	37.2°E	120	ARIX-II
Bruyères-le-Châtel	France	48.9°N	2.3°E	150	SPALAX
Schauinsland	Germany	47.9°N	7.9°E	1,208	SPALAX
Marseille	France	43.2°N	5.2°E	43	SPALAX

Table 2

Overview of the different measurement periods covered in this study

Station	Operational period	Number of valid measurements
Spitsbergen	13 April 2003–31 August 2008	1,798
Stockholm	22 August 2005–31 August 2008	1,540
Dubna	12 November 2006–31 August 2008	1,146
Schauinsland	20 February 2004–31 August 2008	1,165
Bruyères-le-Châtel	8 August 2003–4 July 2005	391
Marseille	29 September 2004–23 March 2005	176

Values below Lc were not reported for the Marseille station

around 18 h. These spectra have been analysed with software from the system developer KRI.

2.3. Time Series Analysis for ^{133}Xe

The data considered in this study were sampled at different time periods, between August 2003 and August 2008. Table 2 gives an overview of the different measurement periods. During this reporting period, not all systems were transmitting data continuously, some of them being used for training operators or undergoing upgrades. The systems in Marseille were only operated for specific factory acceptance test periods.

The xenon isotope most commonly seen in ambient air samples is ^{133}Xe. Its half-life of 5.24 days and its large cumulative fission yields of 6.7 to 7.0% are factors contributing to its high detectability. It is often observed at locations downwind from nuclear power plants and is almost

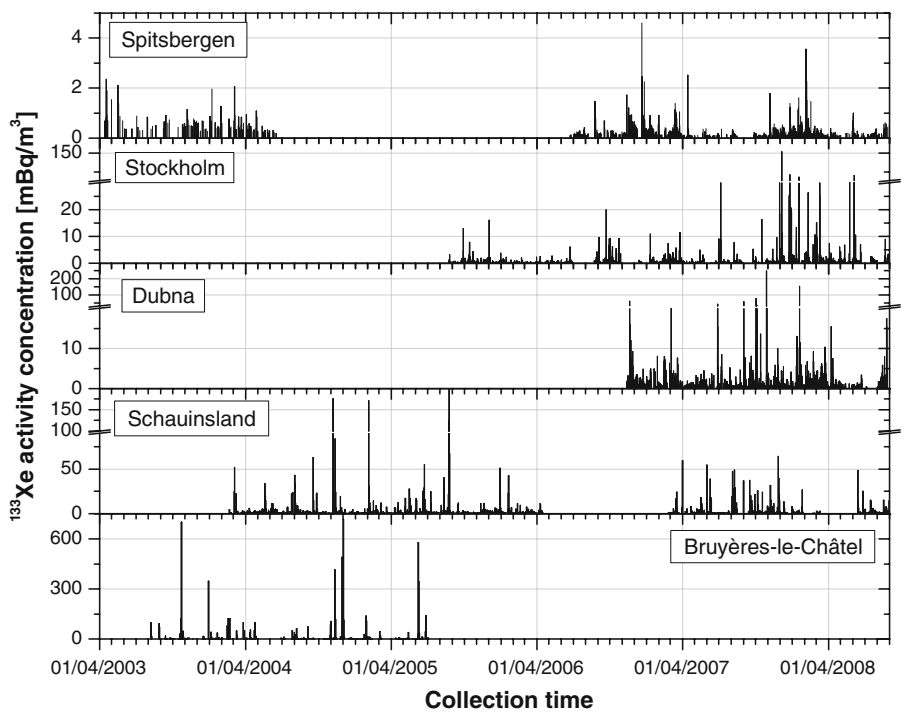

Figure 2
Activity concentration of the ^{133}Xe isotope at the five sites with long time series available. Please note that the plots have different activity concentration scales

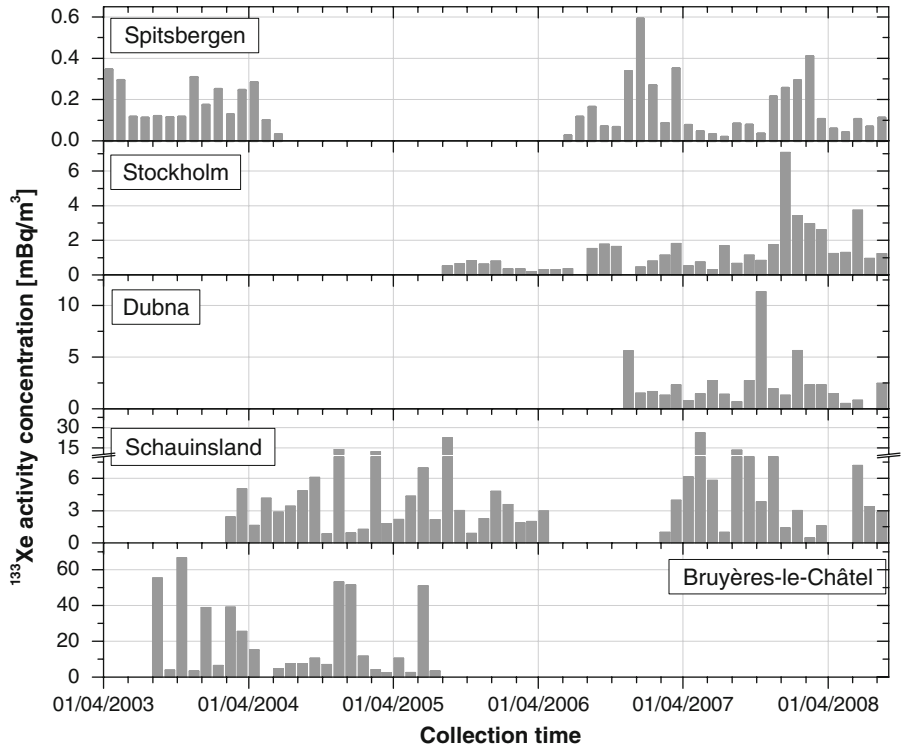

Figure 3
Monthly activity concentrations of ^{133}Xe at five different stations. Please note that the plots have different activity concentration scales

continuously present in the air of the nuclear power dense regions of the globe. Therefore, the main focus of this paper is on this isotope.

Figure 2 presents the activity concentration of all measurements of ^{133}Xe at the five different stations with longer time series. The monthly averages of the activity concentrations are shown in Fig. 3.

At this point it might be useful to note that prevailing activity concentrations of a few mBq/m^3 of ^{133}Xe, as observed in Europe, yields low doses to human beings or other species in the biosphere. The dose rate factor of ^{133}Xe is about 5×10^{-8} mSv/year per mBq/m^3 (KOCHER, 1980). This renders an annual dose to humans of many orders of magnitude less than what is caused by its more natural "cousins" Radon-220 and Radon-222 and their progeny.

The average MDCs of the four different radioxenon isotopes at the six different sites are shown in Table 3. At Marseille, only the MDC from ^{133}Xe was calculated, as the system was under factory

Table 3

The Minimum Detectable Concentration (MDC) and the counting time of the samples at the six stations for the four relevant radioxenon isotopes

Station	^{131m}Xe (mBq/m^3)	^{133m}Xe (mBq/m^3)	^{133}Xe (mBq/m^3)	^{135}Xe (mBq/m^3)	Counting time (h)
Spitsbergen	0.12	0.11	0.19	0.73	11
Stockholm	0.15	0.17	0.23	0.82	11
Dubna	0.3	0.4	0.2	0.9	18
Schauinsland	0.33	1.6	0.12	0.55	23
Bruyères-le-Châtel	6.0	2.0	0.35	0.88	23
Marseille	n.a.	n.a.	0.34	n.a.	23

The MDCs from Bruyères-le-Châtel and Marseille are based on the main gamma peak of the nuclide

acceptance testing. One should note the different counting times of the gas in the detector, which depends on the measurement system used (see Table 1). A shorter counting time increases the time resolution of the sampling but it also decreases the sensitivity. All are, however, still well within the minimum specifications of the IMS (1 mBq/m^3 of ^{133}Xe).

In contrast to observations at the Spitsbergen station at 78°N, that do show higher values in winter than in summer (Saey *et al.*, 2006) there are apparently no such correlations to the season at any of the other stations within the European reactor region. This was also confirmed by Fast Fourier Analyses of the data. We therefore conclude that the release of radioxenon at nuclear facilities in Europe is spread throughout the year whereas Spitsbergen is located remotely enough from xenon emission areas in North America and Europe that long-range atmospheric transport patterns govern the measurements there. These patterns are mainly controlled by the North Atlantic Oscillation that features a pronounced seasonal variability (Saey *et al.*, 2006).

2.4. Background Determination

The distribution of the different activity concentrations of ^{133}Xe at all six different sites is shown in Fig. 4. Measurements below the critical limit (Lc, the value which is used to concretely decide whether a signal is present or not in an actual measurement, with a confidence level of 99.5%) are not plotted but considered as an offset on the probability scale reflecting the fact that a total of 22% of the data were below Lc. The plot suggests that the data are log-normally distributed, which is typical for environmental atmospheric data.

To confirm this distribution, the D'Agostino test (D'Agostino and Stephens, 1986) was performed on the data series of the six different stations. This test can be performed if the data set is larger than 50 and smaller than 1,000 measurements. The non-detects and the extreme values (see Sect. 2.7) were not considered and the significance level α was 0.05. The test was carried out with the values of the activity concentrations and with the logarithmic values of the activity concentrations. Only the test on the logarithmic values confirmed, for all stations except Marseille, the hypothesis of normal distribution which means that the activity concentrations are log-normally distributed.

The frequency distributions, the mean values and the medians of the data sets of ^{133}Xe at the six different sites are shown in Fig. 5.

As five of the data sets were shown to be log-normally distributed, logarithmic scales are used for the activity concentrations. From the sixth, Marseille, very few data points were available. The inflection points in the plots on the higher side indicate optically the change from background to extreme

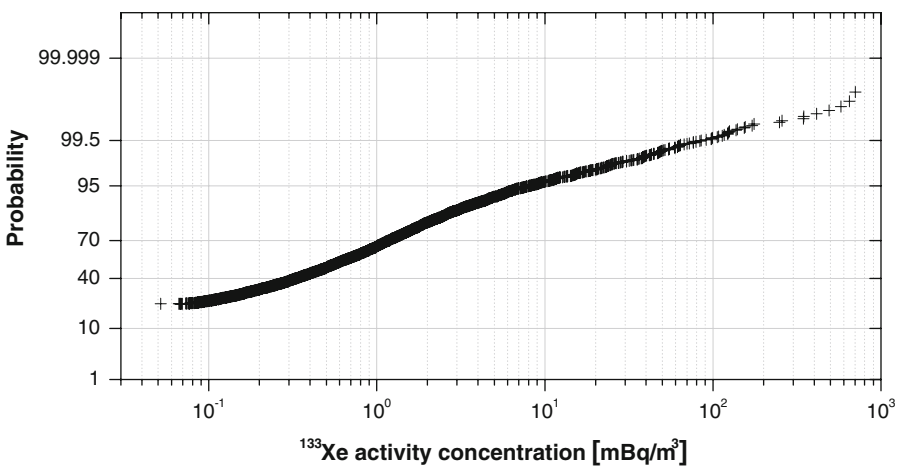

Figure 4
Log-normal Probability Plot of all measurements above the detection limit for all six stations

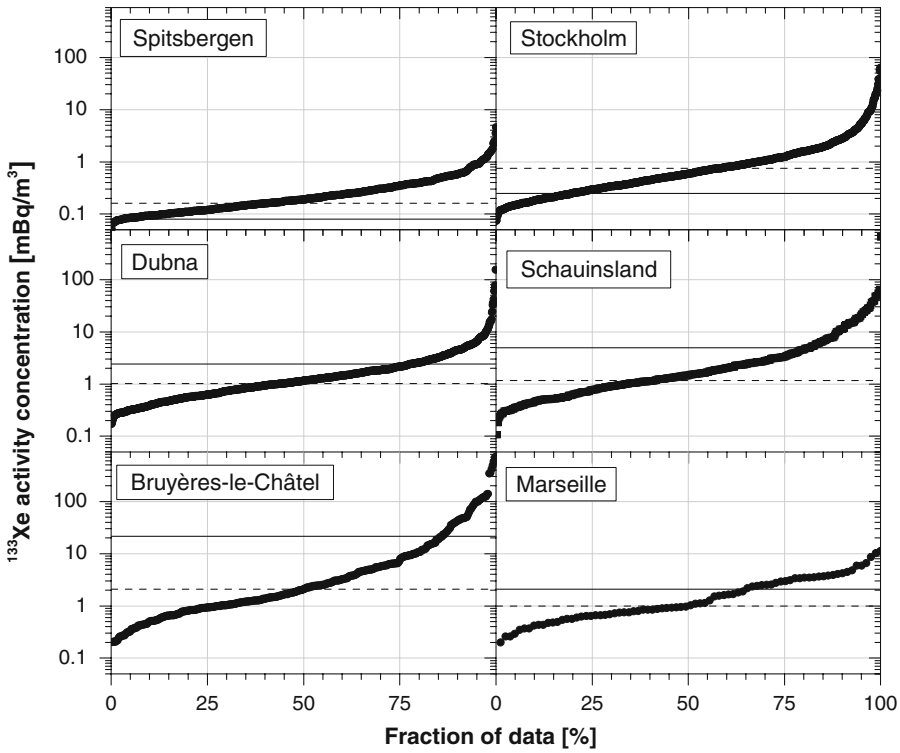

Figure 5
The frequency distribution of ^{133}Xe at the six different sites. The *solid horizontal line* shows the mean value and the *dashed line* the median of the data sets. Data below the critical limit Lc were not plotted

Table 4

Summary of the statistics of the daily ^{133}Xe measurements at the six radioxenon measurement stations presented

Station	Measurements below Lc (%)	Max. (mBq/m^3)	Mean (mBq/m^3)	Median (mBq/m^3)	IQR (mBq/m^3)	75% percentile (mBq/m^3)	95% percentile (mBq/m^3)
Spitsbergen	49	4.61	0.16	0.08	0.25	0.20	0.59
Stockholm	20	155.6	1.39	0.40	0.86	1.01	4.24
Dubna	8.7	249.0	2.43	1.03	1.5	1.97	6.17
Schauinsland	0.18	257	4.3	1.2	2.2	2.8	18.2
Bruyères-le-Châtel	0	717.5	21.4	2.1	5.7	8.0	98.2
Marseille	n.a.	11.1	2.1	1.0	2.3	3.0	5.9

values. Figure 5 shows this in more detail. The Box-and-Whisker diagram of the data set is discussed in Sect. 2.7.

A summary of some statistical parameters is given in Table 4. The values were calculated from all data sets including measurements below Lc. The percentage of measurements below Lc ranges between 0.18% in Schauinsland and 49% in Spitsbergen. For the latter site this means that the median is given by the typical Lc value. The mean has been calculated assuming a zero true value for all measurements below Lc. It should be pointed out that this leads to a systematic underestimation of the true mean, particularly for sites with a high percentage of data points below Lc. For measurements from the Marseille site, no data below Lc were available. In this case the values refer to data above Lc only and both mean and median may be systematically overestimated.

2.5. *Comparison with a Previous Measurement Campaign in Germany*

In the year 2000, an intercomparison exercise with the four different INGE radioxenon measurement systems took place at the BfS in Freiburg, Germany. The goal was to demonstrate the available capabilities and determine the technical characteristics of these new technologies in an independent laboratory away from the developers but still with experienced staff present. The BfS in Freiburg operates a noble gas laboratory and has the required experience in noble gas monitoring to validate the systems by performing re-analysis of the archived samples. This intercomparison exercise is thoroughly described in BOWYER *et al.* (2002) and AUER *et al.* (2004).

The ARSA (Automated Radioxenon Sampler and Analyzer) system from the Pacific Northwest National Laboratory (PNNL, USA), which, like the SAUNA system, was based on beta-gamma coincidence spectrometry, measured the most complete time series of the exercise. Therefore, these data were used to compare the results from the year 2000 with those from 2004 to 2008. The data from 2000 for the isotope ^{133}Xe are presented in Fig. 6. They are based on 874 eight-hour measurements over a period of 340 days (coverage of 86%).

The distribution of this isotope measured in the year 2000 in Freiburg as compared to nearby Schauinsland in the current study, illustrates that there was little change: the most frequently measured activity concentration interval was in both periods the one between 0.4 and 0.8 mBq/m^3.

The frequency distribution has the same shape for both periods. However, the mean and median in the year 2000 (2.31 and 0.80 mBq/m^3, respectively) were lower than in the period 2004–2008 (4.3 and 1.23 mBq/m^3, respectively). This is in contrast to the published data of releases of noble gases from nuclear power plants within the European Community (VAN DER STRICHT and JANSSENS, 2005): in the year 2000 a total airborne release of 9.7 PBq is

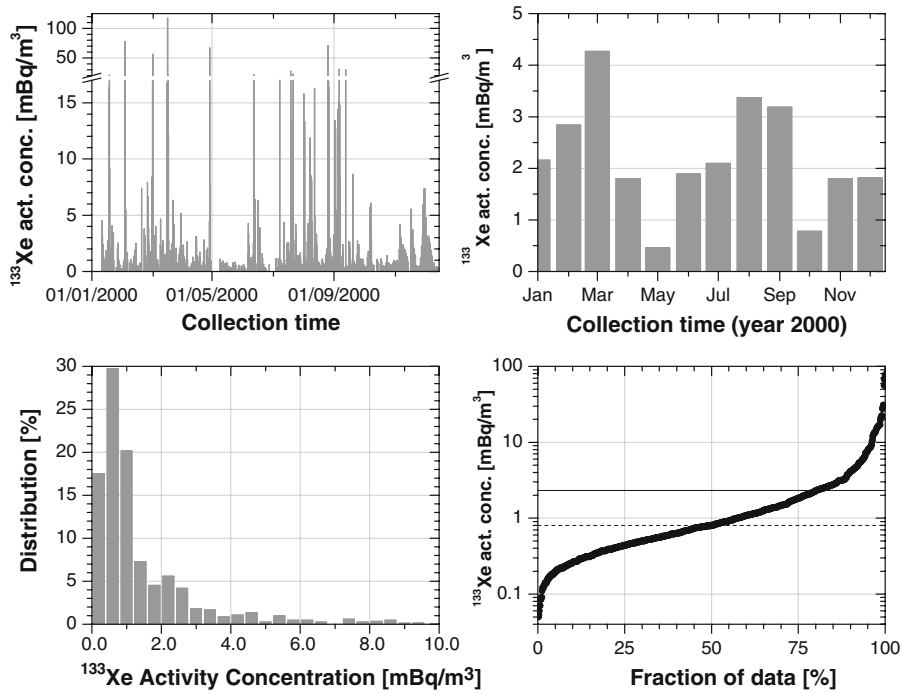

Figure 6
^{133}Xe data from the ARSA system in Freiburg, Germany in 2000. The *upper left graph* shows the activity concentrations of ^{133}Xe. The monthly average activity concentrations are shown in the *upper right graph*. The lower left graph shows the distribution of the activity concentration of ^{133}Xe. The frequency distribution of ^{133}Xe is depicted in the *lower right graph* (the *solid horizontal line* shows the mean value and the *dashed line* the median of the data sets)

reported and 4.7 PBq in 2003, with a generally decreasing trend.

A plausible explanation for the higher activity concentrations measured during 2004 to 2008 is an increased production of the radiopharmaceutical facility in Fleurus, South Belgium (AUER *et al.*, 2010). This facility is remote enough from Freiburg and Schauinsland that emissions from there have already been vented into the free troposphere during their transport to the station. The facility in Fleurus is considered to be a major contributor of environmental radioxenon in Europe (SAEY, 2009). The radiopharmaceutical isotope production facility at Chalk River, Canada (ACHIM *et al.*, 2007) could also impact the detection level in Western Europe.

2.6. Comparison with a Field Campaign in Austria

During July to September 2006 a comprehensive test was conducted in Seibersdorf, Austria, focusing on equipment and procedures to collect and analyse radioxenon samples from the atmosphere and from sub-surface gas. During the test mobile versions of the SAUNA and ARIX systems were deployed. Though the test focussed on operational issues and logistics, 16 atmospheric samples and five sub-surface gas samples were collected and analysed. The activity concentration of ^{133}Xe in most of the atmospheric samples was between 0.3 and 2.4 mBq/m^3, though on one occasion ^{133}Xe activity concentrations of (17 ± 1) and (51 ± 3) mBq/m^3 were reported by the SAUNA and the ARIX system, respectively (AXELSSON, 2007).

2.7. Tests for Extreme Values

Figure 7 shows the Box-and-Whisker diagram of ^{133}Xe measurements at the six different sites. The length of the central boxes indicates the spread of the central 50% of the data. This interquartile range (IQR) in which 50% of the ranked data are found, describes the dispersion of the measurements. It refers only to the data set with data above Lc. Therefore the values differ from those given in Table 4.

The length of the whiskers indicates the extent that the measurements are spread out below and above the central 50% box. The upper whisker extends to the 95th percentile. The – symbol indicates the largest observations and the × symbols show the 99th percentiles. Data between the – and the × are considered to be extreme values. The □ symbol

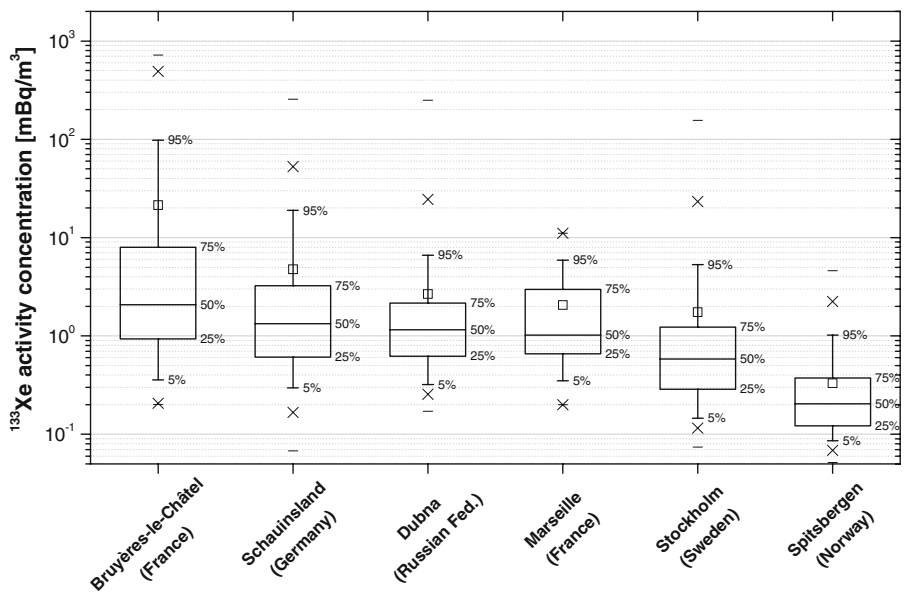

Figure 7
Box-and-Whisker plot of ^{133}Xe at the different sites—values below Lc were not considered here. For details, see text

shows the mean value of the measurements. The lower – symbol indicates the average Lc. In this diagram we can see that the data from the more central European stations Bruyères-le-Châtel and Schauinsland measure generally higher ^{133}Xe activity concentrations than the stations situated on the fringe of the main nuclear power plant region.

The extreme values can be indications of erroneous analyses, but are most probably just the results of extreme emissions at a certain reactor or at a radiopharmaceutical isotope production facility (Saey, 2009). In the framework of the CTBT verification, an extreme value could trigger a special meteorological analysis to identify its possible source region.

2.8. Atmospheric Transport Modelling

2.8.1 Assumption of Sources

The station of Bruyères-le-Châtel is situated in a region with a high density of nuclear power plants—there are roundly 60 operational reactors within a 500-km range. In a 500 km radius surrounding Schauinsland, there are approximately 70 reactors. France produced in the year 2000 half of all nuclear-generated electricity in Europe—Germany almost 20% and Sweden 8% (Energy Information Administration, Office of Coal, Nuclear, Electric and Alternate Fuels, International Nuclear Model, PC Version—May, 2001). Both stations also have Fleurus, the world's third largest radiopharmaceutical isotope production factory (after Chalk River in Canada and Petten in the Netherlands) within the 500-km range. Petten, the world's second biggest producer, has long delay lines installed, which reduce their emission several orders of magnitudes (Saey, 2009). The other two stations have fewer reactors within a 500-km radius: Marseille 15, Stockholm 12 and Spitsbergen none.

Table 5

Order of magnitude of releases of radioxenon at different nuclear facilities

Type of release	Typical order of magnitude of radioxenon release
Hospitals	$\sim 10^6$ Bq/d
Nuclear power plants	$\sim 10^9$–10^{11} Bq/d
Radiopharmaceutical facilities	$\sim 10^{11}$–10^{13} Bq/d
1 kton nuclear explosion underground	0–10^{15} Bq
1 kton nuclear explosion atmospheric	$\sim 10^{16}$ Bq

Table 5 presents an overview of the order of magnitude of radioxenon releases at different types of nuclear installations (UNSCEAR, 2000; Van der Stricht and Janssens, 2005; Kalinowski and Pistner, 2006; Saey, 2007, 2009; Kalinowski and Tuma, 2009).

Some studies concerning the identifications and contributions of the main radioxenon sources in Western Europe (Achim *et al.*, 2007; Le Petit *et al.*, 2008) showed that most of the observed detections at Marseilles, Bruyères-le-Châtel and Schauinsland could be for a first order of approximation, accounted as releases from the European nuclear power plants and from the radioisotope production facility in Fleurus, Belgium.

2.8.2 Modelling of Two Selected Events with the Web-Grape Software

The PTS utilizes Atmospheric Transport Modelling (ATM) to estimate in which regions of the globe a surface level emission into the atmosphere would possibly result in radionuclide detection at one of the sampling stations. Based on the routine global wind field analysis assimilated by the European Centre for Medium range Weather Forecasts (ECMWF), backward atmospheric transport calculations are done for every station and every measurement taken in the network (Wotawa *et al.*, 2003; Becker *et al.*, 2007). The system is organized in a four-layered workflow where the results from the basic ATM calculations are stored as so-called Source Receptor Sensitivity (SRS) fields at a suitable geo-temporal resolution (currently 1°; 3 h) half-way to the workflow.

These fields are then available for post-processing, both in automated mode and in a more flexible interactive mode. In automated mode standard map products called Field-of-Regards (FOR) are generated and included in the daily issued reviewed radionuclide reports. For the interactive mode the client software, Web-Grape, has been developed (Becker, 2006). One source region estimation product that can be generated by Web-Grape in addition to the FOR is the Possible Source Region (PSR),

which is a map that depicts the area or areas that best fit detections in several samples that can be assumed to belong to the same event. In contrast to the measurement specific FOR, the PSR is event-specific and belongs to a scenario of measurements. Web-Grape also features an event calculator (ECAL) with which the user can test different source hypotheses and see at what stations and in what time periods these emissions would lead to detections in the PTS radionuclide network.

Two interesting events were selected as examples to show how a source of an anomalous detection can be located. These examples demonstrate that a regular (daily) sensitivity calculation for each station (i.e., how sensitive a station on a certain day is towards known radioxenon emitting sources), as proposed by UNGAR *et al.* (2007), might help in automatically screening out certain events measured at noble gas stations, i.e., classifying certain high values as resulting from civil sources, as the possible source might be known.

2.8.2.1 One station measurements that could point to the release of a radiopharmaceutical isotope production facility, using one radioxenon isotope The Schauinsland mountain station measures a few times per year activity concentrations above 50 mBq/m^3 of ^{133}Xe, which can be considered as extreme values. Most backtracking calculations with Web-Grape could show that the air that contained those higher radioxenon activity concentrations had passed the south of Belgium 1 or 2 days before. This is an area that hosts the "Institut National des Radioéléments" (IRE) which produces ^{99}Mo for ^{99m}Tc generators and sells its by-product ^{133}Xe to "MDS Nordion SA" located on the same premises near the little town of Fleurus some 40 km south of Brussels. A certain amount of the radioxenon activities produced in this process is released into the atmosphere (SAEY, 2009). As shown in Table 5, these releases can be 10^{11}–10^{13} Bq/day.

Figure 8 displays a backtracking example. The lower left image shows the FOR belonging to the detection of 15 mBq/m^3 of ^{133}Xe in air collected between 14 July 2008 0600 hours and 15 July 2008 0600 hours at the station Schauinsland in Germany (all times in this paper are in UTC). The sensitivity of this measurement (receptor) to all sources on 13 July between 9–12 h is colour-coded and indicates the area where the air sampled during this single measurement at Schauinsland was located at this time. The lower right image shows the PSR of the four measurements taken at the same station between collection start 13 July and collection stop 17 July 2008. There is a confined area where the PSR exhibits values between 0.9 and 1.0. These correlation coefficient values indicate a high consistency of a singular

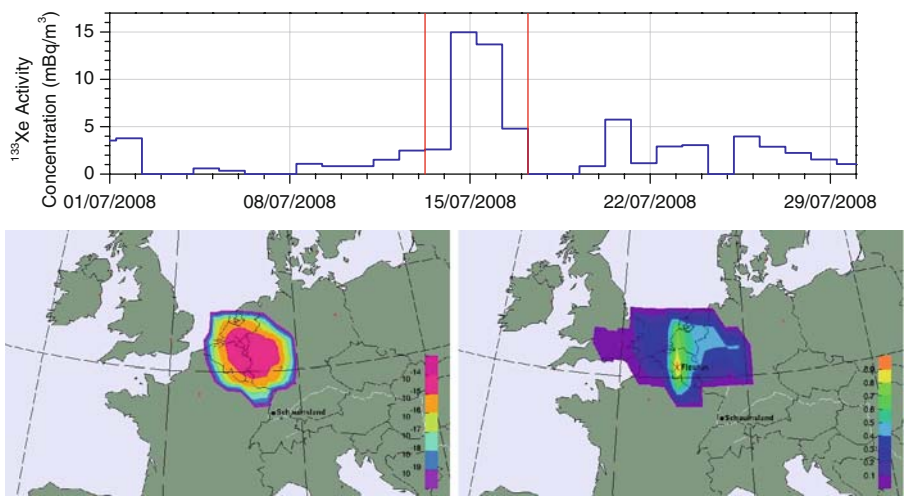

Figure 8
Time series (*upper graph*) of ^{133}Xe for this station for the considered time period. The values between the *red vertical lines* are those used for the PSR calculations. FOR (*lower left*) and PSR (*lower right*) for an interesting measurement of ^{133}Xe at the INGE station on Schauinsland Mountain, Germany

source from there with the four measurements encountered. It is worth noting that the PSR pattern, which utilises more data than the FOR, is more confined than the FOR and corresponds to the region around Fleurus in the south of Belgium. Applying the simple source-receptor relationship equation, Source strength [Bq] × Sensitivity [1/m^3] = Observation [Bq/m^3], the FOR across that area reveals information on the strength of the release: The relation to the release activity is for this area 10^{-15}. The measured activity was 15 mBq/m^3 of ^{133}Xe. Thus it can be concluded that the source should have released around 15×10^{12} Bq on 13 July 2008 between 0900 and 1200 hours. This estimation is consistent with the daily releases from this facility.

2.8.2.2 Two station measurements that could indicate different releases at one radiopharmaceutical facility, using two isotopes Another case was studied in which during the same period (mid-November 2004) in both the Schauinsland Mountain station and the Bruyères-le-Châtel station extreme ^{133}Xe values were measured, 83 and 400 mBq/m^3, respectively (see Fig. 9). The ^{135}Xe isotope was also present in these samples.

Figure 9 shows the isotopic activity ratio of ^{135}Xe/^{133}Xe of the samples collected between 11 and 13 November 2004. They are positioned on one of the parallel lines which has the slope of the radioactive decay of the ratio. This could indicate the hypothesis that they are related to the same emission.

The sample with collection stop on 6 November 2004 is clearly related to another source and the 13 November sample is not fully consistent with the cluster of four samples and will, therefore, not be considered further.

Initial atmospheric transport modelling indicates that the samples measured in Bruyères-le-Châtel and Schauinsland might originate from the same sources: the radiopharmaceutical isotope production facility in Fleurus. However, meteorological backtracking analysis reveals that both stations did not see the same

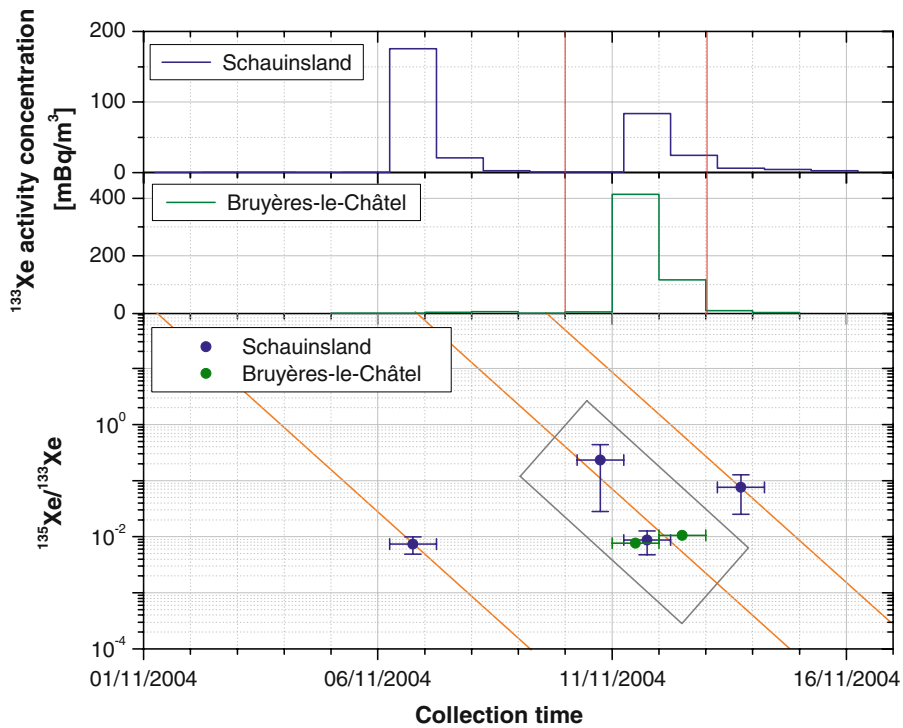

Figure 9
Time series (upper two graphs) of ^{133}Xe for Schauinsland and Bruyères-le-Châtel for the considered time period. The values between the *red vertical lines* are those used for the PSR calculations. The *lower graph* shows the ^{135}Xe/^{133}Xe ratio in early November 2004. The *orange lines* have slopes corresponding to the ratio decay. The four samples in the *box* may be associated with the same event in accordance with radioactive decay, but only ATM shows that it is not the case. The *horizontal bars* indicate the 24-h sampling time

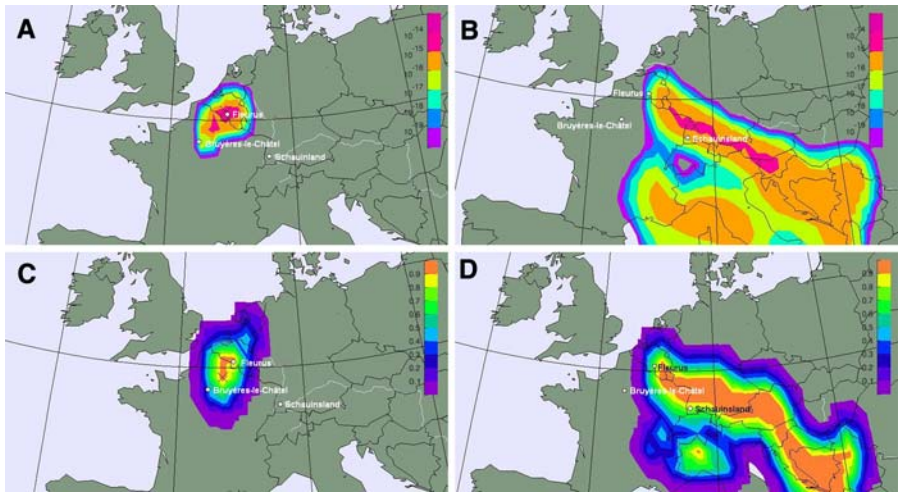

Figure 10
FOR (*upper two images*) and PSR (*lower two images*) for an extreme value measurement of ^{133}Xe at the INGE station in Bruyères-le-Châtel, France (*two left images*) and on Schauinsland Mountain, Germany (*two right images*), for the time period indicated in Fig. 9—A detailed description is given in the text

release. The facility in Fleurus released in that period daily. The samples measured in Schauinsland originate from a release that took place on 9 November (see right-hand side FOR and PSR plots for 3–6 UTC of Fig. 10) while the samples measured in Bruyères-le-Châtel most likely originate from a release 2 days later, on 11 November, 3–6 UTC (see left-hand side FOR and PSR plots of Fig. 10). This result has been verified by forward atmospheric modelling, assuming these release times at Fleurus yield a very good correlation between the observed and modelled concentrations at Bruyères-le-Châtel and Schauinsland of 0.94 and 0.96, respectively, however the latter only while applying a 1-d phase correction in view of the excessively flat representation of the topography in the wind-field model running at 1° horizontal resolution.

Additionally other sources were checked. During that period, there was only one nuclear power plant that went through a shut-down or start-up operation, according to reports from the IAEA Power Reactor Information System (PRIS). This was a reactor start-up operation at Gravelines, France, situated at the sea side near the Belgium border, after a refuelling on 8 November. According to the ATM calculations, the release should have been six orders of magnitude above the average nuclear power plant releases to explain the measurements and can, therefore, be excluded.

This example illustrates that simultaneous detections at different stations and measurements of multiple isotopes can point to a possible source. It also demonstrates the crucial role of ATM in source identification.

3. Summary and Outlook

Long-term continuous data from four radioxenon isotopes at six different European radioxenon stations have been analysed:

- Three stations are situated in the middle of the European nuclear facilities (nuclear power plants and two radiopharmaceutical isotope production facilities), two are just outside this zone and one is a remote, polar station. The measurements show that the releases of radioxenon at nuclear facilities in Europe are spread throughout the year;
- The radio xenon background at several stations in the European network, including variations in time and geographical location, was well characterized during recent years. This is a very valuable data set for further developing a categorisation scheme to discriminate releases from civil sources against releases from nuclear tests;
- A comparison between ^{133}Xe data from the year 2000 and the years 2003 to 2008 has shown that

there was a slight increase in the radioxenon activity concentrations in the Freiburg area, which might originate from an increase in radiopharmaceutical production of ^{99}Mo of around 5% per year;
- The background values for these nuclides at each of these stations have been determined—the atmospheric activity concentrations were log-normal distributed. The mean dose rate has been calculated to be 9 orders of magnitude less than the one from the common present ^{222}Rn gas;
- Some selected examples have been used to demonstrate how meteorological backtracking analyses can be used to indicate the possible source locations and source strengths that would account for the observations.

To reach a routine for an extreme value flagging system which could be used to automatically screen out extreme values, the following topics would need more careful study:

- release scenarios from different types of nuclear facilities (theoretical and experimental);
- automatic sensitivity calculations of the INGE stations towards all known major radioxenon emitters;
- field measurements around these facilities to better understand and quantify the initial dilution of batch releases.

An early information system of the nuclear facilities themselves regarding significant releases would help build confidence in the measurements.

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References

Aatami (2003), *User Manual of Radionuclide Analysis and Evaluation Software Aatami*, Version 3.15. Preparatory Commission of the Comprehensive Nuclear Test Ban Treaty Organisation, Vienna, Austria.

Achim, P., Bellivier, A., Armand, P., Taffary, T., Fontaine, J.P., and Piwowarczyk, J., C. (2007), *Contributions of xenon releases in the atmosphere from radionuclide production facilities and nuclear nuclear power plants to the detection of ^{133}Xe by SPALAX systems in werstern Europe*, Proc. 11th Internat. Conf. *Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes*.

Auer, M., Kumberg, T., Sartorius, H., Wernsperger, B., and Schlosser, C. (2010), *Ten years of development of equipment for measurement of atmospheric radioactive xenon for the verification of the CTBT,* Pure Appl. Geophys. *167*, 4–5.

Auer, M., Axelsson, A., Blanchard, X., Bowyer, T.W., Brachet, G., Bulowski, I., Dubasov, Y., Elmgren, K., Fontaine, J.P., Harms, W., Hayes, J.C., Heimbigner, T.R., McIntyre, J.I., Panisko, M.E., Popov, Y., Ringbom, A., Sartorius, H., Schmid, S., Schulze, J., Schlosser, C., Taffary, T., Weiss, W., and Wernsperger, B. (2004), *Intercomparison experiments of systems for the measurement of xenon radionuclides in the atmosphere,* Appl. Radiation and Isotopes *60*, 6, 863–877.

Axelsson, A. (2007), *Development, Demonstration, Testing and Evaluation of On-Site Inspection Equipment for Xenon Sampling*, Separation and Measurement. CTBTO Technical Report, CTBT/PTS/TR/2007-1, CTBTO, Vienna.

Becker, A. (2006), *A new tool for NDC analysis of atmospheric transport calculations,* CTBTO Spectrum *7*, 19–24.

Becker, A., Wotawa, G., De Geer, L.-E., Seibert, P., Draxler, R.R., Sloan, C., D'Amours, R., Hort, M., Glaab, H., Heinrich, P., Grillon, Y., Shershakov, V., Katayama, K., Zhang, Y., Stewart, P., Hirtl, M., Jean, M., and Chen, P. (2007), *Global backtracking of anthropogenic radionuclides by means of a receptor oriented ensemble dispersion modelling system in support of Nuclear-Test-Ban Treaty verification,* Atmos. Environ. *41*, 21, 4520–4534.

Bernström, B. and De Geer, L.-E. (1983), *Mätning av Små Mängder Xenon-133 I Luft,* FOA-rapport ISSN 0347-3694, C 20515-A1, FOA, Stockholm, Sweden.

Bieringer, J. and Schlosser, C. (2004), *Monitoring ground-level air for trace analysis: Methods and results,* Analyt. Bioanalyt. Chem. *379*, 2, 234–241.

BMU (2007), *Umweltradioaktivität und Strahlenbelastung—Jahresbericht 2006* (in German), Reaktorsicherheit, Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit, pp. 100–104.

Bowyer, T.W., Schlosser, C., Abel, K.H., Auer, M., Hayes, J.C., Heimbigner, T.R., McIntyre, J.I., Panisko, M.E., Reeder, P.L., Satorius, H., Schulze, J., and Weiss, W. (2002), *Detection and analysis of xenon isotopes for the comprehensive nuclear-test-ban treaty international monitoring system,* J. Environ. Radioact. *59*, 2, 139–151.

D'Agostino, R.B. and Stephens, M.A. (1986), *Goodness-of-Fit Techniques,* (Marcel Dekker) 0824774876.

Dahlman, O., Mykkeltveit, S., and Haak H. (2009), *Nuclear Test Ban: Converting Political Visions to Reality* (Springer, 1st edition, 2009), ISBN-13: 978–1402068836.

De Geer, L.-E. (1996), *Atmospheric radionuclide monitoring: A Swedish perspective*. In Huseby, E.S. and Dainty, A.M., eds., *Monitoring a Comprehensive Nuclear Test Ban Treaty* (pp. 157–177) (The Netherlands: Kluwer Academic Publishers, 1996).

Dubasov, Y.V., Popov, Y.S., Prelovskii, V.V., Donets, A.Y., Kazarinov, N.M., V. V. Mishurinskii, Popov, V.Y., Rykov, Y.M., and Skirda, N.V. (2005), *The АРИК-01 Automatic facility for measuring concentrations of radioactive xenon isotopes in the atmosphere,* Instrum. Experim. Techn. *48*, 3, 373–379.

Ehhalt, D., Muennich, K.O., Roether, W., Schoelch, J., and Stich, W. (1963), *Artificially produced radioactive noble gases in the atmosphere,* J. Geophys. Res. *68*, 13, 3817–3821.

Fontaine, J.P., Pointurier, F., Blanchard, X., and Taffary, T. (2004), *Atmospheric xenon radioactive isotope monitoring,* J. Environ. Radioact. *72*, 1–2, 129–135.

Igarashi, Y., Miyao, T., Aoyama, M., Hirose, K., Sartorius, H., and Weiss, W. (2000a), *Radioactive noble gases in surface air monitored at MRI, Tsukuba, before and after the JCO accident,* J. Environ. Radioact. *50*, 1–2, 107–118.

Igarashi, Y., Sartorius, H., Miyao, T., Weiss, W., Fushimi, K., Aoyama, M., Hirose, K., and Inoue, H.Y. (2000b), *^{85}Kr and ^{133}Xe monitoring at MRI, Tsukuba and its importance,* J. Environ. Radioact. *48*, 2, 191–202.

Kalinowski, M.B. and Pistner, C. (2006), *Isotopic signature of atmospheric xenon released from light water reactors,* J. Environ. Radioact. *88*, 3, 215–235.

Kalinowski, M.B. and Tuma, M.P. (2009), *Global radioxenon emission inventory based on nuclear power reactor reports,* J. Environ. Radioact. *100*, 58–70.

Kalinowski, M.B., Axelsson, A., Bean, M., Blanchard, X., Bowyer, T.W., Brachet, G., McIntyre, J.I., Peters, J., Pistner, C., Raith, M., Ringbom, A., Saey, P.R.J., Schlosser, C., Stocki, T.J., Taffary, T., and Ungar, R.K. (2010), *Discrimination of nuclear explosions against civilian sources based on atmospheric xenon isotopic activity ratios,* Pure Appl. Geophys. *167*, 4–5.

Kocher, D.C. (1980), *Dose-rate conversion factors for external exposure to photon and electron radiation from radionuclides occurring in routine releases from nuclear fuel cycle facilities,* Health Phys. *38*, 543.

Le Petit, G., Armand, P., Brachet, G., Taffary, T., Fontaine, J., P., Achim, P., Blanchard, X., Piwowarczyk, J. C., and Pointurier, F. (2008), *Contribution to the development of atmospheric radioxenon monitoring*, J. Radioanalyt. Nuclear Chem. *276*, 2, 391–398.

Mikhailov, V., Dubasov, Y.V., and Matushenko, A.M. (1999), *Nuclear Explosions in the USSR, The North Test Site, Reference Material on nuclear explosions, radiology, radiation safety (ver. 2)*. Tests, Interagency Expert Commission on Assessment of Radiation and Seismic Safety of Underground Nuclear, V.G. Khlopin Radium Institute, Russian Nuclear Society, St. Petersburg, Russian Federation.

Pakhomov, S.A. and Dubasov, Y.V. (2008). *Estimation of explosion energy yield at Chernobyl NPP accident*, Geophys. Res. Abstr. *10*, A-09280.

Ringbom, A., Larson, T., Axelsson, A., Elmgren, K., and Johansson, C. (2003), *SAUNA–a system for automatic sampling, processing, and analysis of radioactive xenon,* Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, *508*, 3, 542–553.

Saey, P.R.J. (2007), *Ultra-low-level measurements of argon, krypton and radioxenon for treaty verification purposes,* ESARDA Bull. *36*, July 2007, 42–55.

Saey, P.R.J. (2009), *The influence of radiopharmaceutical isotope production on the global radioxenon background,* J. Environ. Radioact. *100*, 5, 396–406.

Saey, P.R.J. and De Geer, L.-E. (2005), *Notes on radioxenon measurements for CTBT verification purposes,* Appl. Radiation Isotopes *63*, 5–6, 765–773.

Saey, P.R.J., Wotawa, G., De Geer, L.-E., Axelsson, A., Bean, M., d'Amours, R., Elmgren, K., Peterson, J., Ringbom, A., Stocki, T.J., and Ungar, R.K. (2006), *Radioxenon background at high northern latitudes,* J. Geophys. Res. *111*, D17306.

Schlosser, C., Sartorius, H., and Schmid, S. (2003), *Status of the SPALAX at the RN-33 station,* International Workshop on Atmospheric Radioxenon Measurements, Ottawa, Canada, 29 September–2 October, 2003.

Schölch, J., Stich, W., and Münnich, K.O. (1966), *Measurements of radioactive xenon in the atmosphere,* Tellus *XVIII*, 2, 298–300.

Stockburger, H., Sartorius, H., and Sittkus, A. (1977), *Messung der Krypton-85- und Xenon-133-Aktivität der atmosphärischen Luft*., Z. Naturforsch. *32a*, 1249–1253.

UNGA (1996), *United Nations General Assembly Resolution Number 50/245*. In U.N. New York, U.S.A.

Ungar, K., Hoffman, I., Yi, J., Stocki, T., De Geer, L.-E., Walters, M., and Becker, A. (2007), *Use of existing IDC routine numerical calculations as a prototype standard radionuclide numerical ATM product for operational categorization and categorization development*. In *International Noble Gas Experiment, Workshop,* Las Vegas, NV, U.S.A.

UNSCEAR (2000), *Sources and Effects of Ionizing Radiation*, Report to the General Assembly by the United Nations Scientific Committee on the Effects of Atomic Radiation. 1, Nations, United, UNSCEAR, New York.

Van der Stricht, S., and Janssens, A. (2005), *Radioactive effluents from nuclear power stations and nuclear fuel reprocessing plants in the European Union, 1999–2003*. Radiation Protection 143, Directorate-General Environment.

Weiss, W. and Leeb, H. (1993), IMIS—The German integrated radioactivity information and decision support system, Radiat. Prot. Dosimetry *50*, 2–4, 163–170.

Wotawa, G., De Geer, L.-E., Denier, P., Kalinowski, M., Toivonen, H., D'Amours, R., Desiato, F., Issartel, J.-P., Langer, M., Seibert, P., Frank, A., Sloan, C., and Yamazawa, H. (2003), *Atmospheric transport modelling in support of CTBT verification—Overview and basic concepts,* Atmos. Environ., *37*, 18, 2529–2537.

Ziegler, C.A. and Jacobson, D. (1995), *Spying Without Spies: Origins of America's Secret Nuclear Surveillance System,* Westport, CT, USA (Praeger Publishers 1995), ISBN-13: 978-0275950491.

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