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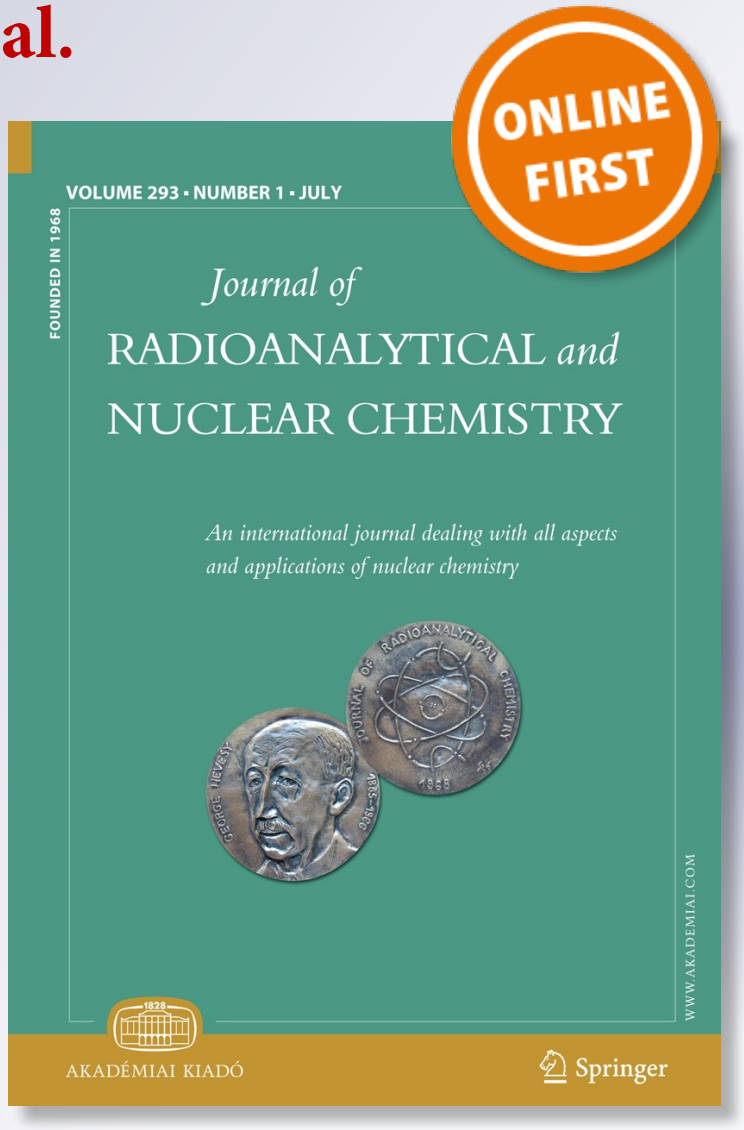
Journal of Radioanalytical and Nuclear Chemistry
An International Journal Dealing with All Aspects and Applications of Nuclear Chemistry

ISSN 0236-5731

J Radioanal Nucl Chem
DOI 10.1007/s10967-012-2025-2



Springer

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Received: 15 July 2012

Abstract The Comprehensive Nuclear-Test-Ban Treaty (CTBT) specifies that radioxenon measurements should be performed at 40 or more stations worldwide within the International Monitoring System (IMS). Measuring radioxenon is one of the principle techniques to detect underground nuclear explosions. Specifically, presence and ratios of different radioxenon isotopes allows determining whether a detection event under consideration originated from a nuclear explosion or a civilian source. However, radioxenon monitoring on a global scale is a novel technology and the global civil background must be characterized sufficiently. This paper lays out a study, based on several unique measurement campaigns, of the worldwide concentrations and sources of verification relevant xenon isotopes. It complements the experience already gathered with radioxenon measurements within the CTBT IMS programme and focuses on locations in Belgium, Germany, Kuwait, Thailand and South Africa where very little information was available on ambient xenon levels or interesting sites offered opportunities to learn more about emissions from known sources. The findings corroborate the hypothesis that a few major radioxenon sources contribute in great part to the global radioxenon background. Additionally, the existence of independent sources of ^{131m}Xe (the daughter of ^{131}I) has been demonstrated, which has some potential to bias the isotopic signature of signals from nuclear explosions.

Keywords Radioxenon · Treaty verification · Medical isotope production facilities · Nuclear power plant

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Published online: 08 August 2012

Introduction

Radioxenon and its role in CTBT verification

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 crucial for the pursuit of nuclear disarmament: it bans any kind of nuclear explosion.

Several verification techniques are provided. Beside wave form measurements (seismic, hydro-acoustic and infra-sound), radionuclides in ground level air are measured (radioactive particulates and noble gases) and atmospheric transport modelling (ATM) is used to connect treaty relevant radionuclide or noble gas detections to a possible source region and, if possible, to a waveform detection. When the International Monitoring System (IMS) network is completed, there will be 337 stations worldwide; among them 80 measuring permanently radionuclide particles where at least 40 of these will also be equipped with noble gas measurement systems [5, 20].

The radioisotopes of the noble gas xenon play a major role when confirming whether or not an underground explosion was nuclear in nature [6]. In addition to nuclear explosions, radioxenon isotopes can also be created in, among others, nuclear power plants, research reactors or medical isotope production facilities [14]. To distinguish the radioxenon coming from a nuclear explosion from other environmental sources, the noble gas background (absolute activity concentration and isotopic composition) has to be studied carefully and the sources have to be well understood.

International noble gas experiment

In the beginning of the implementation of the International Monitoring System, noble gas monitoring was not a mature technology and few data were available on the global radioxenon background. Continuous radioxenon measurement systems had to be developed, as no commercial systems were available. Four countries, France, Russia, Sweden and USA, all with experience of atmospheric xenon measurements, offered to develop such systems. To test the developed noble gas systems, the Provisional Technical Secretariat (PTS) for the CTBT Organisation (CTBTO) and the developers work together in the International Noble Gas Experiment (INGE) project [1].

These systems are now being installed at several worldwide clusters and send their results online to the International Data Centre (IDC) in Vienna for analysis. Currently, 29 stations are operational or in construction phase, equivalent to more than 70 % of the projected number at entry into force of the Treaty. However, some regions on the globe are not covered yet.

The announced nuclear test by the Democratic Peoples Republic of Korea in October 2006 not only highlighted the importance of early entry into force of the Treaty, it also underscored the need for the rapid build-up of the CTBT verification regime [3, 13, 15]. The event constituted a real life system-wide test for the PTS and highlighted the potential value that the global verification system can bring to States Signatories. It demonstrated the technical relevance or the CTBT verification arrangement in general and especially the unique role of noble gas monitoring, as well as the need to get a good understanding of the global noble gas background.

Worldwide noble gas background

Currently INGE stations are collecting data in the North- and South-Americas, Europe [17, 18], North-Asia and Oceania. In order to ensure the ability to characterize the global emissions as seen in the current and future IMS's noble gas measurement network, it is of vital importance to know the "noble gas background". An essential part of the data analysis, categorization of events detected by noble gas system, needs to be developed. This is especially important in regions of the world where currently no stations exist. Noble gas field measurements are the best way to achieve this and to provide answers to such "unknowns". These field campaigns can also perform measurements at different distances around possible sources like e.g. medical isotope facilities, which IMS stations can't.

The "EU/JA-II" project

It has now been shown that xenon emissions from anthropogenic sources could hide relevant radioxenon signals from nuclear explosions [18]. As described among others in Saey [16] medical isotope production facilities (MIPF) were identified to be the main source with possible similar ratio signatures as explosions, but no high-sensitive measurements from the stack of these facilities or close to these main emitters had ever been performed to confirm the numerical studies.

Besides having numerical simulations and some long distance measurements, it is important to compare this numerical knowledge with measurements very close to the MIPF sources, and if possible even directly from the releasing stack, before the released gas gets diluted to very low levels and gets mixed with air that might contain radioxenon isotopes from other sources.

Therefore, in 2008 and 2009 two field campaigns in and around medical isotope production facilities were

conducted. One took place in and around the MIPF of Fleurus, Belgium and a second one in and around the MIPF in Pelindaba, South Africa. Further, two field campaigns took place in areas were there were no IMS noble gas stations but nuclear facilities in a broader area. The places that were identified and where there was also a logistical support available were Safat in Kuwait, Cape Point in South Africa and Chiang Mai in Thailand.

The campaigns were carried out as a joint collaboration between the CTBTO, the Swedish Defence Research Agency (FOI) and Pacific Northwest National Laboratory (PNNL) and were primarily sponsored by the Council of the European Union (EU) in the framework of the implementation of the EU Strategy against the Proliferation of Weapons of Mass Destruction (the second EU Joint Action for supporting the CTBTO).

SAUNA measurement system

These different measurement campaigns used the Swedish SAUNA-II (Swedish Automatic Unit for Noble gas Acquisition). This sampling and measurement equipment uses beta-gamma coincidence spectrometry [12]. It has already been used in several field campaigns and was thus well evaluated [13]. The system delivers data with detection limits similar to the IMS version for the four relevant noble gas isotopes (^{131m}Xe, ^{133m}Xe, ^{133}Xe and ^{135}Xe) and the data are therefore directly applicable to the IMS. The equipment needs only a covered and air-conditioned area of around 6 m × 3 m and a floor that can take a load of 700 kg/m^2. The required power is maximum 8 kW (50 Hz, 380 V three phases, ground plus neutral).

Field measurements

Campaign around the Fleurus (Belgium) MIPF

During the summer of 2008, a field campaign in the region around the Belgian MIPF in Fleurus, the National Institute for Radioelements (IRE), which is the world's third largest one, was performed at distances between 0.5 and 100 km from the stack (see Fig. 1), the two largest being MDS Nordion in Chalk River, Canada and Tyco Healthcare in Petten, The Netherlands [9]. The scope was to obtain high-sensitive radioxenon measurements of the releases and the isotopic composition during and after production activities in the factory.

Systematic "plume hunting" (measuring at a site where it was predicted the day before that the plume should pass) was performed during 3 weeks using a mobile SAUNA sampler in combination with meteorological forecasts. Further, the facility allowed exceptionally to take three samples directly from the stack itself. All the samples were transported daily from Belgium to Sweden where they were measured with a SAUNA laboratory system. In total 35 atmospheric and 3 stack samples from this campaign were measured at the FOI noble gas laboratory in Stockholm.

It was found that the atmospheric ^{133}Xe concentrations were in the range 0.7–4 × 10^5 mBq/m^3. The stack samples had concentrations in the range 2 × 10^9–4 × 10^{10} mBq/m^3—the flow rate of that stack is 100,000 m^3/h. This is compatible with the reported total release of around 5 × 10^{12} Bq/day. Nine of the 35 atmospheric samples collected had detectable concentrations of all four relevant xenon isotopes.

Fig. 1 The Belgian campaign's sampling sites—left the area close to the Fleurus facility, right the further away sampling places

Campaign around the Pelindaba MIPF in South Africa

The world's fourth largest radiopharmaceutical facility, NTP Radioisotopes Ltd, is located near Pretoria, in Pelindaba, South Africa. The only other facilities that are known to emit any radioxenon on the African continent south of the Equator are a small nuclear power plant (Koeberg), located 1,300 km southwest, near Cape Town and a small research reactor in Kinshasa (D.R. of Congo), located 2,700 km northwest. This means that the MIPF source is dominant with respect to xenon emission in the region.

Between 10 November and 22 December 2008, atmospheric radioxenon was measured continuously at the North-West University, Mafikeng, South Africa, which is situated 250 km west of Pelindaba, at the border with Botswana. The activity concentration of the measured isotopes ^{133m}Xe, ^{133}Xe and ^{135}Xe is shown in Fig. 2.

In parallel, stack samples were taken at the NTP facility on an almost daily basis and measured with a HPGe gamma detector at the local laboratory of South African Nuclear Energy Corporation (NECSA).

During this 1 month of operation in Mafikeng, the measured radioxenon content of the sampled air varied between zero and 27 mBq/m^3 with a mean value of 3.24 mBq/m^3 for ^{133}Xe. Using ATM, the relatively high peak values have been identified to be due to releases from the Pelindaba MIPF (see Fig. 3).

Campaign in Cape Point, South Africa

Following the Mafikeng campaign, the SAUNA-II xenon sampling and measurement system equipment was shipped to Cape Point, 50 km south of Cape Town and 1,300 km southwest from the Pelindaba facility. Between 16 January and 2 February 2009, radioxenon was measured continuously.

During these 3 weeks of operation, no measurable amounts of any of the four relevant radioxenon isotopes were observed. Figure 4 shows a summation of all sampled spectra from both detectors of the SAUNA system. The arrows indicate known gamma peaks from radon daughters, but it was determined that no appreciable radioxenon could be detected. The 30-keV xenon X-ray region, where the X-rays from the xenon isotopes 131 m, 133 m, and 133 were expected, is indicated with a red arrow—no peak was found here either.

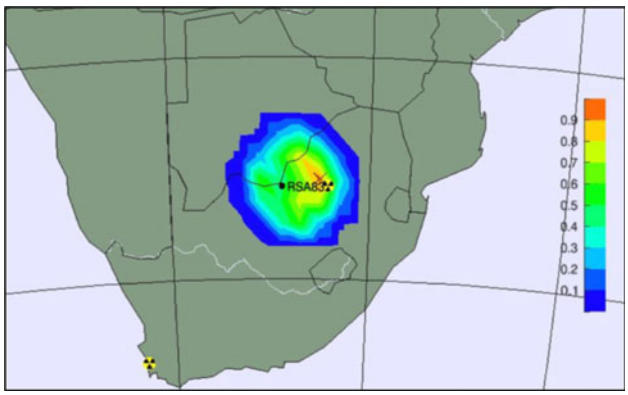

Fig. 3 The Possible Source Region for all the three measurements from 2–3 December 2008 (the *scale* indicates the correlation coefficient values—a high value means a high consistency of a single source from there with the five measurements encountered). The *nuclear symbol* indicates the NPP near Cape Town and the RIPF in Pelindaba. The calculations were made using the software Web-Grape [4]

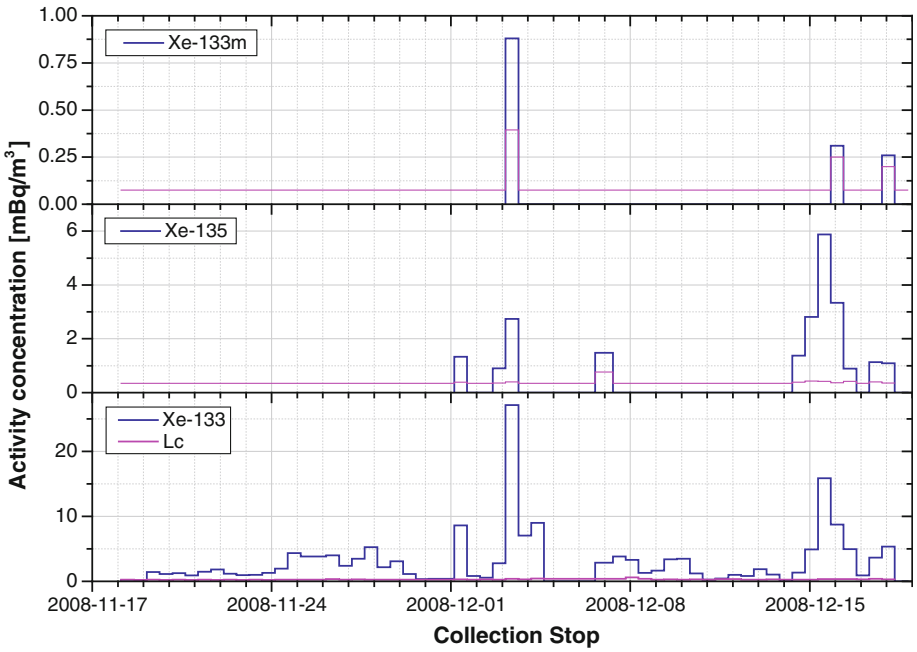

Fig. 2 Activity concentrations (*blue*) and critical detection limits (*red*) of the three measured radioxenon isotopes at the Mafikeng site during the above described measurement campaign. (Color figure online)

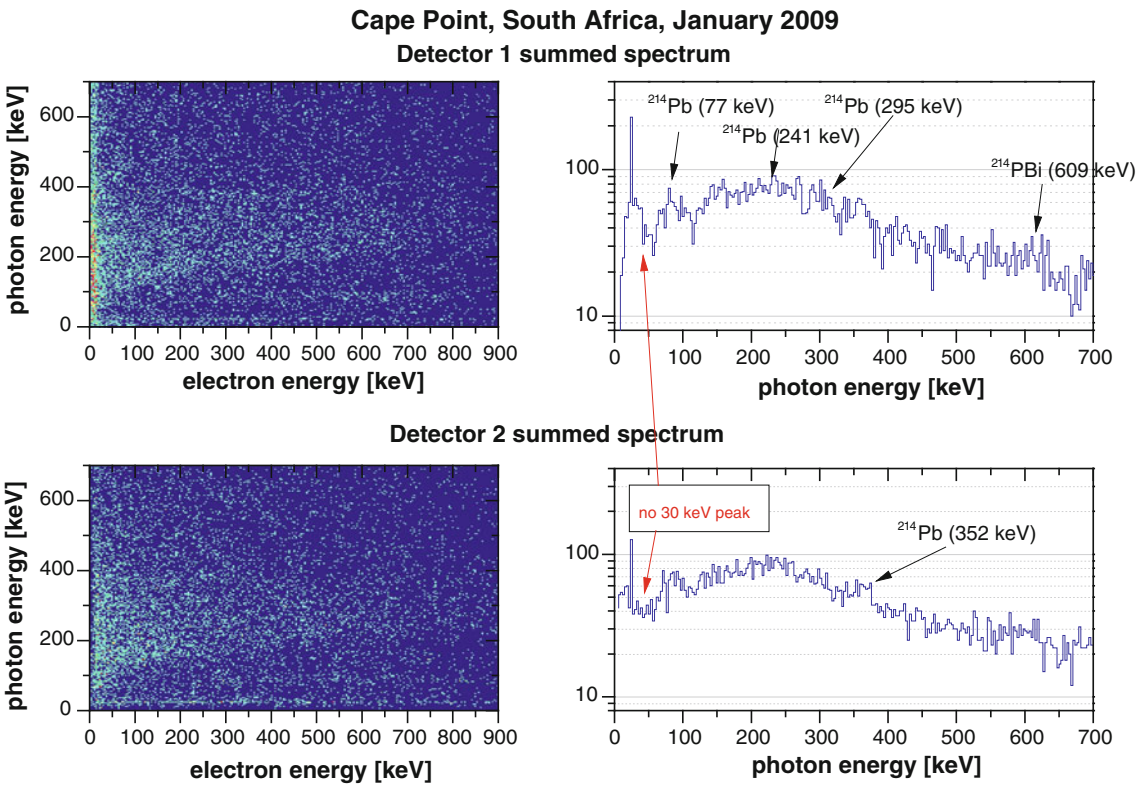

Fig. 4 Beta-gamma and gamma spectra (in coincidence with beta) for the summed sample spectra measured by detector 1 (*upper* two graphs) and detector 2 (*lower* two graphs) in Cape Point between 16 January and 2 February 2009

Atmospheric transport modelling showed that the air measured during this campaign originated from above the Southern Atlantic Ocean and from Antarctica.

Campaign in Safat, Kuwait

Kuwait is located in the north-east corner of the Arabian Peninsula; it is a flat country and covered mostly by the sandy Arabian Desert.

The Environment and Urban Development Division of the Kuwait Institute for Scientific Research (KISR) in Safat, near Kuwait City, had agreed to host the experiment. The building in which the existing IMS particulate monitoring system from the RASA type (IMS station name: KWP40) is installed, was selected to host the equipment as it provided enough space for the SAUNA noble gas measuring system and a climate-controlled environment (see Fig. 5).

The system produced 113 samples with a stable xenon volume greater than 0.3 ml. In general, the radioxenon levels are low in Kuwait—low but clearly detected ^{133}Xe levels were measured in over 40 samples and ^{131m}Xe was detected on at least five occasions (see Figs. 6, 9). No clear detections of ^{133m}Xe or ^{135}Xe were observed during the measurement campaign. The highest observed ^{133}Xe level was 0.83 $\pm$ 0.11 mBq/m^3 and the mean concentration was 0.17 mBq/m^3. The observed ^{133}Xe plumes were broad, ranging over several days or a week, indicating that the source must have been very remote.

Campaign in Chiang Mai, Thailand

Between 22 February and 30 April 2009, radioxenon was measured continuously at the Plasma and Beam Physics Research Facility, Chiang Mai University.

The measurement results (see Fig. 7) show clear indications of radioxenon, although on a low level: a maximum of 0.57 of ^{133}Xe mBq/m^3 and a maximum of 0.41 of ^{131m}Xe mBq/m^3 were measured during the campaign.

The fact that Chiang Mai exhibits such low concentrations is significant. The conditions for radioxenon measurements were not known before these measurements and since little in known about local sources, these measurements were important to make.

Campaign near a nuclear power plant in Germany

A field campaign took place between 2 and 8 March 2009 at the Isar-I nuclear power plant (NPP) during a scheduled revision after 18 months of operation. The goal of this campaign was to assess the direct impact of NPP's on radioxenon background, capturing the exceptional event of a reactor shutdown with direct releases bypassing the

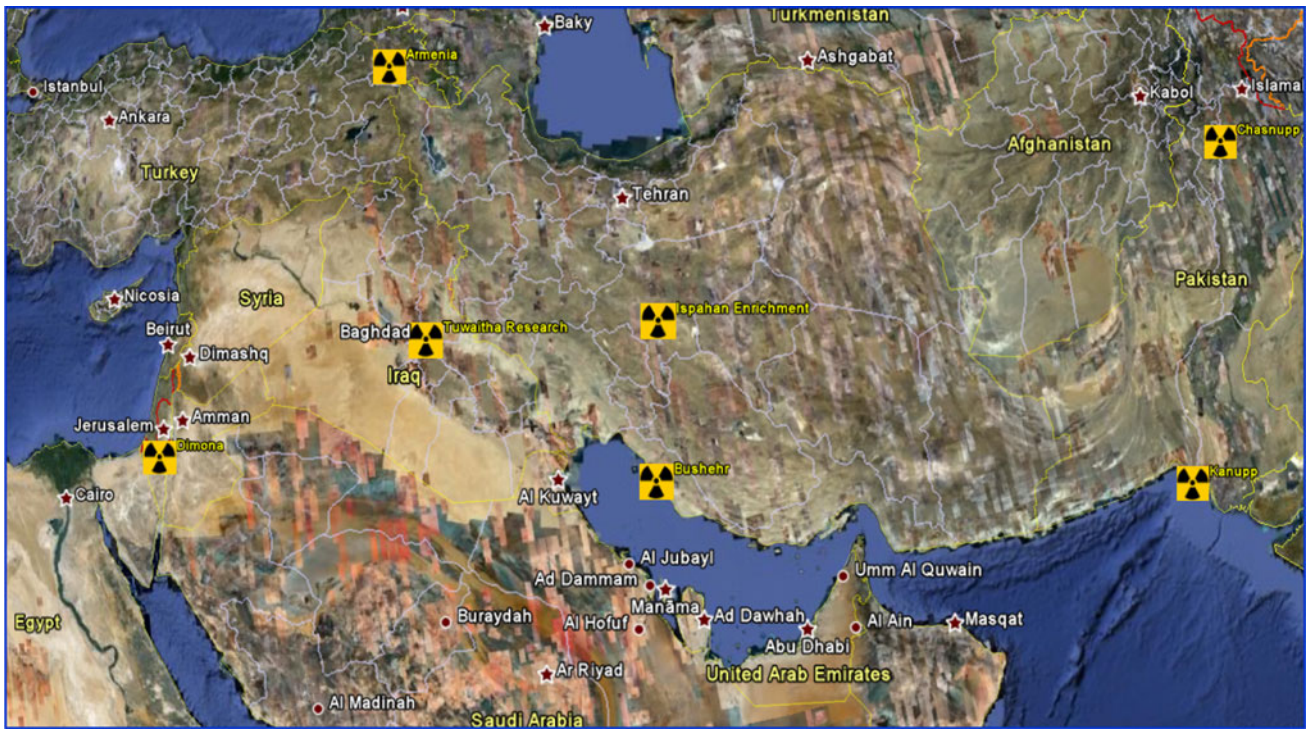

Fig. 5 Map of the Middle East, centered around Kuwait. The *nuclear symbols* indicate the NPP in the area. The measurement campaign in Kuwait started on 5 September and ended on 1 December 2008. During operations, there were large amounts of sand in the fan filters of the SAUNA, which required a regular filter cleaning

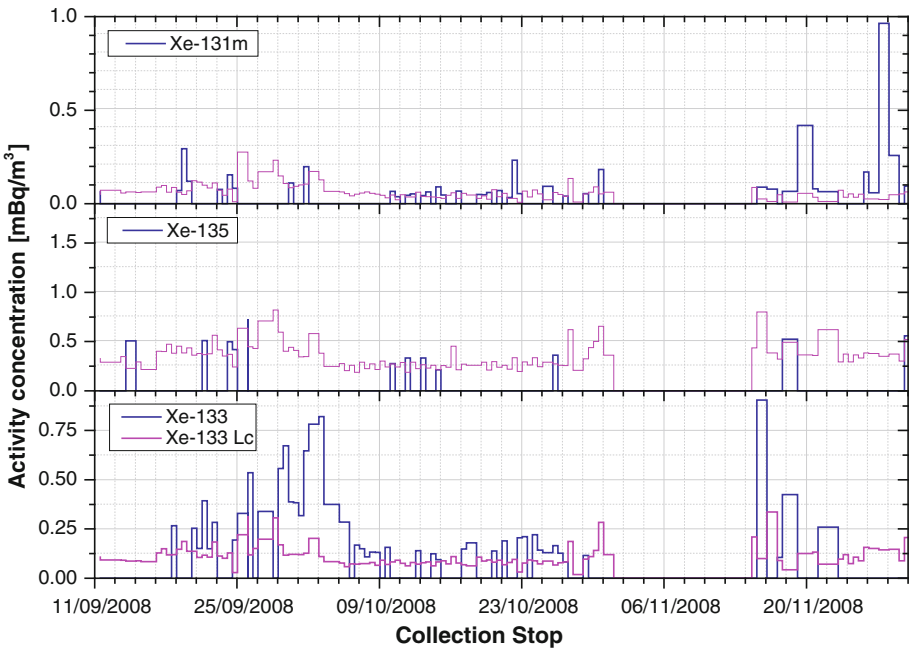

Fig. 6 Activity concentrations (*blue*) and critical detection limits (*red*) time series of the radioxenon isotopes 131 m, 133 and 135 measured in Safat, Kuwait. (Color figure online)

charcoal decay assembly. The Isar-I NPP is a boiling-water reactor with a thermal power of 878 MW that was commissioned in 1977.

FOI brought and used their mobile sampling equipment, which was mounted on a truck. It was operated similar as the Fleurus campaign (see section "Campaign around the Fleurus (Belgium) MIPF"). The team went sampling according to meteorological forecasts made by the German Bundesamt für Strahlenschutz (BfS), who also measured the collected samples using a SAUNA II laboratory system. On 4–5 March four background samples were taken. According to the plant, there was a very small pre-release

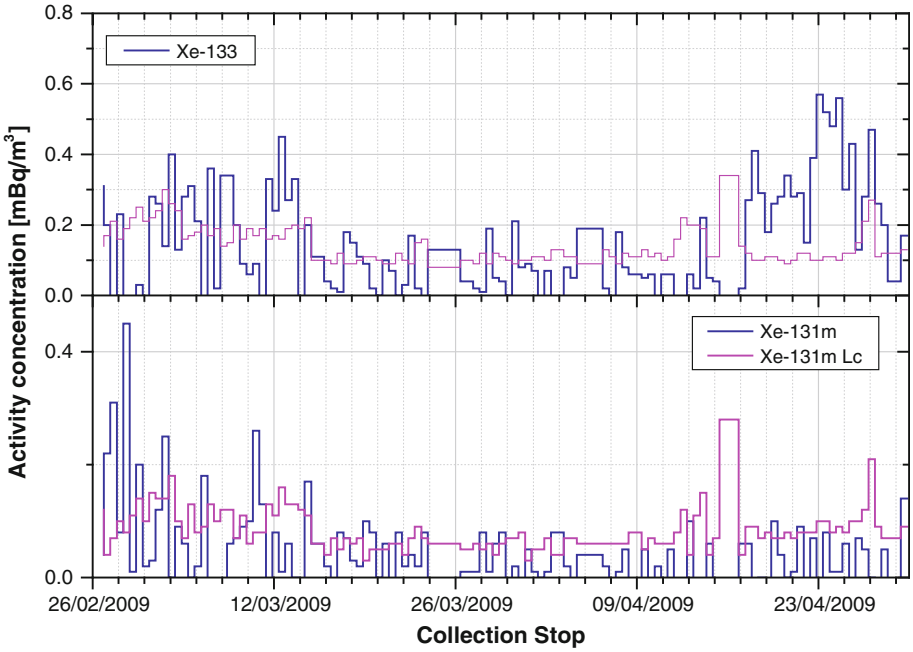

Fig. 7 Time series of the radioxenon isotopes 131 m and 133 in Chiang Mai, Thailand

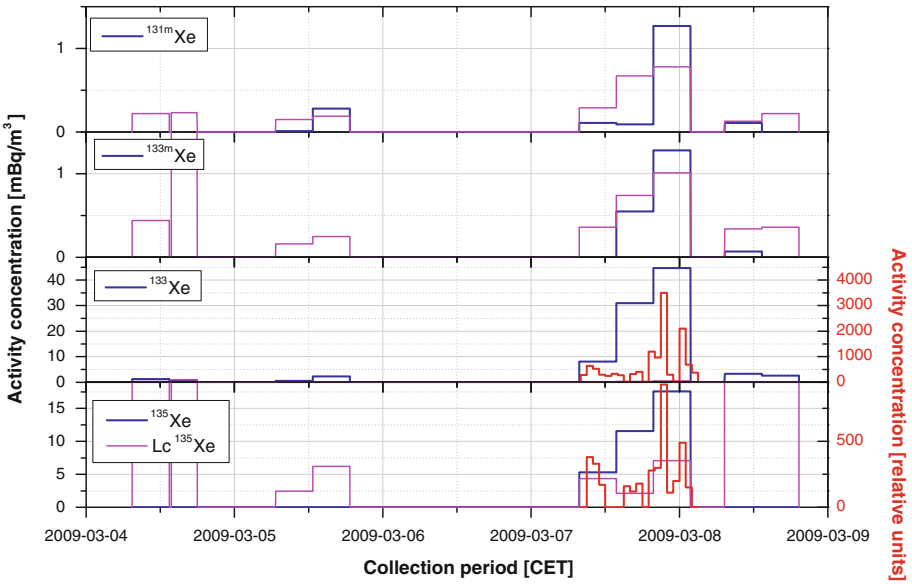

Fig. 8 Measured activity concentrations of the CTBT relevant radioxenon isotopes from air collected in the vicinity of the Isar-I nuclear power plant (*left axis*) and the reported stack emission data (*right axis*)

on March 6 between 16:00 and 18:00 h and the full release started on March 7 at 8:00 h and ended on March 8 at 3:00 h (all times CET). The results from the measured air samples are shown in Fig. 8.

Results and discussion

Radioxenon from RIPF's

The measurement campaigns in the Fleurus (Belgium) and Pelindaba (South Africa) area show the need to have a good understanding of the local conditions, both in terms of potential sources nearby and of meteorology. The stack measurements at both facilities correspond to a daily release of around 5×10^{12}–1×10^{13} Bq. This is consistent with typical release rates for this type of facility and well below exposure guidelines around the facility and thus not dangerous to the public. On the other hand it is expected to be high enough to increase the radioxenon background in wide regions around the facility and has a potential impact on the monitoring capability of the highly sensitive CTBT xenon monitoring systems.

Results of a comparison of the observed isotopic ratios in the air surrounding the Fleurus and Pelindaba facilities were published earlier [17]. The calculations made with

tools described in [11] and [21], using data from [7], show a very good correspondence with the environmental measurements for the Pelindaba facility. At the IRE facility, however, there are several measurements that are not corresponding with the calculated ratios. A likely explanation is the several other activities that take place in that facility. It was reported in personal communications that beside the batch releases after a ^{99}Mo production, there are other independent releases in the same stack from e.g. ^{131}I production, from cleaning activities in the hot cells, etc.

When atmospheric radioxenon is measured, it is of key importance to understand the isotopical mix and the process of its origin. The mechanism for gas emissions from facilities into the atmosphere can vary greatly with different processes in different facilities, different laboratories in a facility and the different durations the different noble gases will stay in certain parts of the facility. At several facilities there is additional production of ^{131}I via the ^{130}Te(n,p)^{131}I process of which 1.2 % decays to ^{131m}Xe.

Most facilities use the Cintichem process, a method developed by Union Carbide in 1968 at their Cintichem facility in which the uranium targets are dissolved in nitric acid [2]. Noble gases and iodine are released together during uranium dissolution. A part will then also enter the stack, decay to ^{131m}Xe and join the already present other xenon isotopes.

Dissolving the uranium/aluminium targets in a basic solution will release the noble fission gases from the dissolver solution, but not the iodine. In a next step, which can be performed several days later, lowering the pH of the solution will release iodine isotopes into the gas phase, allowing their separate recovery as done by e.g. the MIPF facility in Petten, The Netherlands and by CNEA in Argentina [19].

Radioxenon in low background areas

It was shown before that the radioxenon activity concentration in Kuwait and Thailand were low compared to large areas in Europe and North America.

As can be seen in Fig. 9, ^{133}Xe and ^{131m}Xe are anti-correlated: in 36 samples, ^{133}Xe is present (i.e., value is larger than the critical limit of detection), but no ^{131m}Xe, in 29 samples, ^{131m}Xe is present, but no ^{133}Xe and only in 11 samples both nuclides are present. This might indicate that ^{133}Xe and ^{131m}Xe originate from different sources.

After the campaign, the atmospheric transport models were calculated with the real meteorological data for the time period the campaign took place. It was shown that Kuwait monitored the near and middle east, as well as Northern Africa and later (November) also Europe with a possible extend to the Northern USA and South-East Canada, where the Chalk River Laboratory is situated. The samples which contained mainly ^{133}Xe (beginning of October) are correlated to air-masses from Europe. The lower ^{133}Xe activity concentration samples as well as those containing only ^{131m}Xe were seen mainly at the end of October and they occur when the station is in contact with more local air.

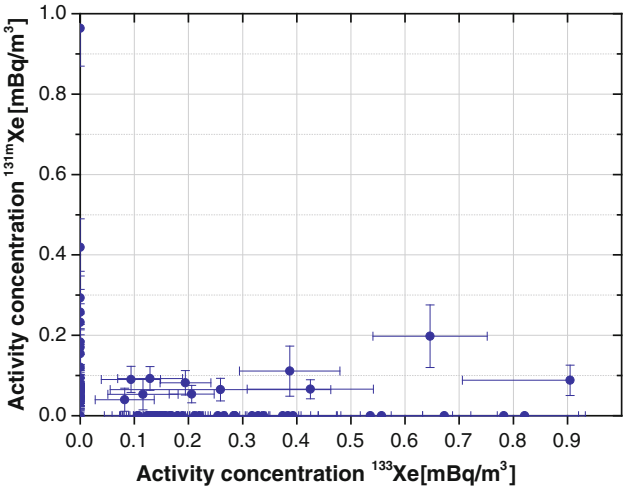

Fig. 9 Correlation between ^{133}Xe and ^{131m}Xe activity concentrations measured in Kuwait

A similar graph was made for the results from the Thai campaign (Fig. 10). In 46 samples, ^{133}Xe is present without ^{131m}Xe; in 22 samples, ^{131m}Xe is present without ^{133}Xe. Both nuclides are present in only five samples. This indicates that ^{133}Xe and ^{131m}Xe also here most probably originate from different sources.

The highest detection of ^{133}Xe measured in Chiang Mai was 0.57 mBq/m^3. Sample collection stopped on April 23

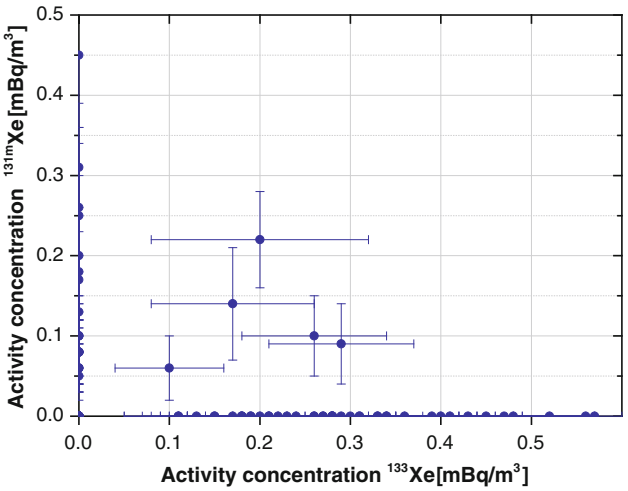

Fig. 10 Correlation between ^{133}Xe and ^{131m}Xe activity concentrations measured in Chiang Mai, Thailand

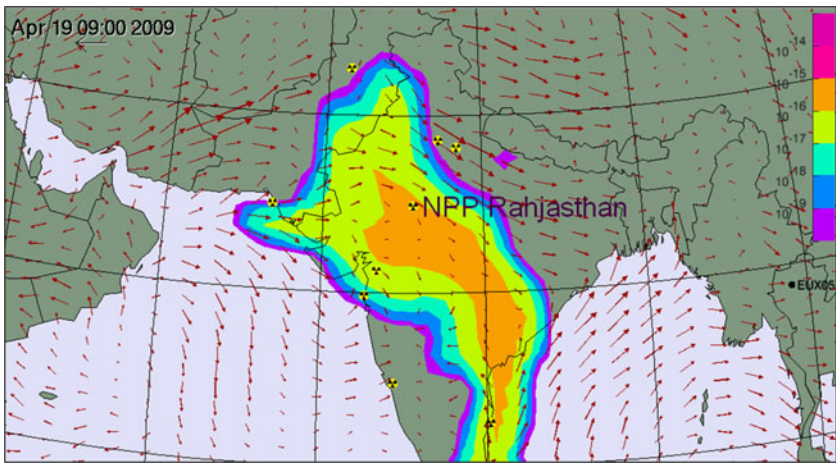

Fig. 11 Backtracking example for the Chiang Mai measurement of April 23 2009

at 03:00 UTC. Identifying a possible source region, using the calculated field of regard for that measurement, shows a possible release from the nuclear power plant of Rajasthan (India) around April 19 or another Indian nuclear power plant (NPP) (Fig. 11). However, the signal was so weak and singular that there is no chance to discriminate one NPP from the other. Measured ^{131m}Xe levels can be easily explained by ^{131}I decay originating from medical application in local hospitals.

Environmental radioxenon during a NPP shutdown

As can be seen in Fig. 12, the two measurements of air collected in the vicinity of the Isar-1 plant which contained all four relevant radioxenon isotopes match the isotopic ratios of civil sources, i.e., they are in the ratio graph situated near the steady-state equilibrium point for a Boiling Water Reactor (BWR).

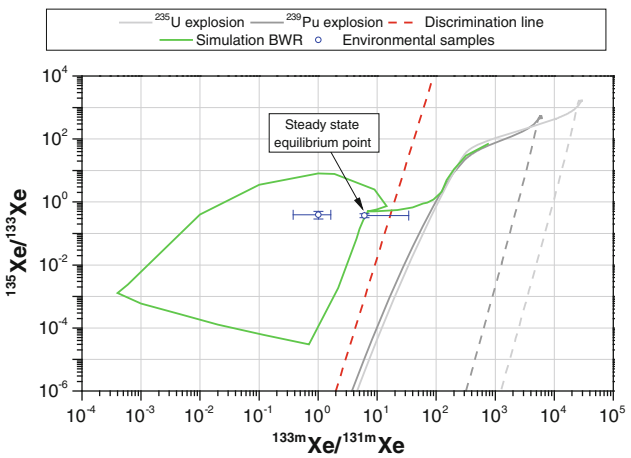

Fig. 12 Four-Isotopic graph indicating the measurements (*blue*), a simulated reactor cycle (*green*), the discrimination line (*red*, see [8]), and two types of nuclear explosions (*grey*). (Color figure online)

Conclusions

Analyses of the performed radioxenon measurement campaign data indicate that knowledge about releases from MIPFs in Belgium (Fleurus) and South Africa (Pelindaba), two of the four largest MIPFs worldwide, is considerably improved. For the first time, off-site radioxenon measurements were taken during the shut-down of a NPP. Together with first measurements of remote areas (Kuwait and Thailand), this project provided valuable progress towards completing a global radioxenon inventory to fill the gaps in areas where no data were yet available.

The findings corroborate the hypothesis that a few major radioxenon sources contribute in great part to the global radioxenon background. A more specific knowledge of source characteristics will be helpful to improve the capabilities of the IMS to distinguish these civil sources from treaty-relevant detections. A reduction of emissions by a factor of 1000 is technically possible for several of these known MIPFs and would bring the releases to the same level as nuclear power plants ($\sim 10^9$ Bq/day). The benefits of such reductions has been communicated accordingly to the radiopharmaceutical-producing community in the framework of the Workshop on Signatures of Medical and Industrial Isotope Production (WOSMIP, see also [10]), especially considering that future medical isotope production is predicted to increase, and mitigation drives the need to understand the chemical processes that allow releases.

Regarding data from the NTP plant in South Africa, the measured ratios match theoretical modelling within a margin of error of 10 %. However, the predictions and measurements of xenon isotopic ratios at the IRE plant are significantly different and indicate that further work is needed to get a better understanding of the processes there.

The project revealed many important findings on environmental radioxenon, which have been highlighted

before. However, the campaigns were too short to fully reveal the seasonal influences and to fully understand phenomena like temporal decoupling, where due to inversions or stagnation local sampled air is decoupled from regional air possibly bearing radioxenon from distant sources. It is recommended, therefore, that such campaigns be continued to gain longer time series of environmental radioxenon.

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