DOI: 10.1007/s10967-008-0517-x Journal of Radioanalytical and Nuclear Chemistry, Vol. 276, No.2 (2008) 391–398

APPLICATION OF NUCLEAR TECHNIQUES TO NATIONAL SECURITY AND TREATY MONITORING

Contribution to the development of atmospheric radioxenon monitoring

G. Le Petit,* P. Armand, G. Brachet, T. Taffary, J. P. Fontaine, P. Achim, X. Blanchard, J. C. Piwowarczyk, F. Pointurier

Commissariat à l'Energie Atomique (CEA), DIF/DASE/SRCE, BP 12, 91680 Bruyères-le-Châtel, France

(Received March 2, 2007)

Within the frame of Comprehensive Nuclear-Test Ban Treaty (CTBT), this paper deals with the development of the new techniques necessary for the xenon monitoring requested by the CTBT. An automatic system called SPALAX™, devoted to the on-site sampling and measurement was developed by French atomic energy commission (CEA). Analytical methods and equipments have been studied at our laboratory, using dual X-γ-spectrometry in order to get independent means with better sensitivity within a robust quality assurance program. In the case of a wide number of potential existing sources and depending on meteorological conditions, several solutions can be arrived at.

Introduction

The monitoring of atmospheric radioactive xenon is one of the technologies used for the detection of nuclear tests. The CTBTO will have to implement a network of 40 radioxenon systems worldwide.[1] In this context CEA developed a high sensitive and automated fieldable system named SPALAX™ which is industrialized and commercialized by our partner (Environnement S.A). This system extracts, purifies and concentrates xenon from the air[2] and measures four radioxenons (^{131m}Xe, ^{133}Xe, ^{133m}Xe, ^{135}Xe) using high resolution gamma-spectrometry. In addition, to support the network, some accredited laboratories will guarantee the quality of the data by analyzing samples coming from the archives of the systems with a high level of sensitivity. A methodology based on mesoscale atmospheric transport modeling has been developed to explain the detections.

Source characterization

Xenon isotopes are relevant signatures of nuclear explosions. However, there are more than 400 nuclear power plants worldwide (around 150 of them are located in the European Union), research reactors, reprocessing plants, medical production and application facilities that release radioactive xenon during normal or incidental operations.

For the verification of compliance with the CTBT, it is important to bring reliable elements allowing the discrimination between nuclear explosion and nuclear civilian activities. It has been expected that the xenon activity ratios would be useful for source discrimination and event timing as well.[3,4] This assertion is based on the fact that a nuclear explosion is an instantaneous process with a very short duration of neutron flux. In a nuclear reactor, the irradiation time is very long with a neutron flux in thermal equilibrium so that isotopic compositions are very different from those generated by a nuclear explosion.

As examples, ^{135}Xe and ^{133m}Xe are important components in a nuclear explosion and the ratio of ^{135}Xe/^{133}Xe just after the test is several orders of magnitude larger than that of civilian releases and ^{133m}Xe/^{133}Xe ratio is different by 2 orders of magnitude. KALINOWSKI, UNGAR and BOWYER[5] show that the situation is in reality more complex in many cases because the resulting isotopic composition is depends strongly on the reactor neutron flux which varies during shut down and start up periods, and leads to variable ratios. This result was obtained notably by the storage of the gas and the mixing of fresh and aged air masses. Consequently, the theoretically derived isotopic ratios spread over orders of magnitude which does not allow to draw a unique conclusion in all the cases to discriminate nuclear explosion from civilian activities. It has been necessary to develop a method based on the correlation between the isotopic ratios. It is called Multi Isotopic Ratio Correlation (MIRC) and allows to screen out irrelevant cases. A straight line (Fig. 1) delimits 2 domains likely to discriminate a nuclear test against nuclear civilian activities. This graph has been determined[6] by using data from the Nevada Test Site and from nuclear power plant releases.

* E-mail: gilbert.le-petit@cea.fr

0236–5731/USD 20.00

Akadémiai Kiadó, Budapest
Springer, Dordrecht

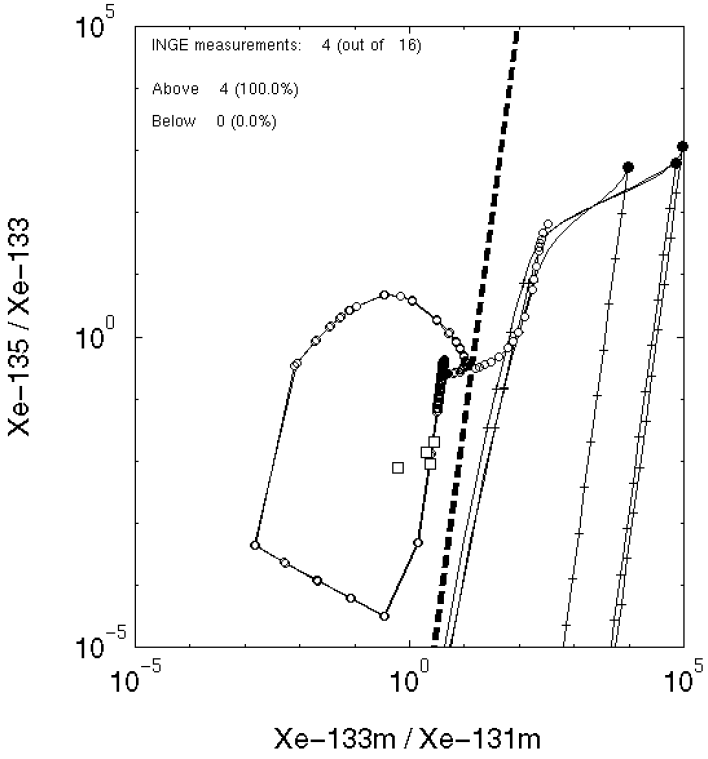

Fig. 1. Source discrimination based on Multi Isotopic Ratio Correlation (MIRC) method

Gas transfer processing from a SPALAX™ archive bottle at the laboratory

We designed a simple gas transfer device dedicated to transfer the SPALAX™ archive bottle contents to the same counting cell used by the SPALAX™ system which will be deployed at our CTBT certified laboratory for verifying and, if needed, supplementing the results of the station. The experimental setup includes a vacuum pump and an automated syringe. The SPALAX™ archive bottle (volume 300 cm^3, pressure 9 kPa) and the counting cell (volume 25 cm^3) are linked to the syringe (volume 450 cm^3); the dead volume which includes the different connectors is estimated to be less than 10 cm^3. The transfer of the xenon from the archive is done by successive steps including compression and pressure reduction until the pressure equilibrium between the archive and the experimental setup dead volume is achieved. Due to a possible xenon loss in the system from the archiving process and a xenon loss inherent to our experimental setup, the transfer yield is estimated to be at about 60%. The transfer time of the process is estimated to be about 90 minutes. At the end of the transfer the counting cell pressure is about 55 kPa.

Measurements of stable xenon volume and air equivalent volume in the archive

The experimental setup used to analyze the stable xenon concentration of an archived sample is based on isothermal gas phase chromatography. The chromatograph is equipped with two 2-metre long analytical columns filled with a grafted polymer and with a thermal conductivity detector (TCD). The adsorbent allows the chromatographic separation of the gas mixture (namely N_2, CO_2 and Xe). By using external gas standards, it is possible to calibrate the thermal conductivity signals, and then to calculate the xenon concentration of the archived sample $[Xe]_S$.

The air volume equivalent to the volume amount of Xe analyzed by X-γ-spectrometry (V_{EQ}) is then given by the following equation:

$$V_{EQ}\ (\mathrm{m}^3 STP) = \frac{V_{Cell}}{0.087}[\mathrm{Xe}]_S \frac{P_{Cell}}{P_0}\frac{T_0}{T_{Cell}}$$

where V_{Cell} is the measurement cell volume, P_{Cell} the gas pressure in the cell, T_{Cell} the gas temperature in the cell, $P_0 = 1013.25 \cdot 10^5\ Pa$, $T_0 = 273.15$ K.

Xenon dual X-γ-spectrometry analysis

The four xenon isotopes likely to be present in a sample can be measured by gamma ray or X-ray analysis, essentially using the $K\alpha_1$ and $K\alpha_2$ rays which exhibit the higher emission probability. The factor of merit defined by:

$$F = \frac{I \cdot \varepsilon_E}{\sqrt{\mu_B}}$$

where I is the γ or X probability emission, ε_E the full energy peak efficiency at the energy E and μ_B the background under the peak, clearly shows (Table 1) that ^{131m}Xe and to a lesser extent ^{133m}Xe, have to be measured from the X-rays in order to achieve the lowest detection limit while ^{133}Xe and ^{135}Xe have to be measured from the gamma-rays.

The detection of the radioxenons is carried out using a high resolution low level spectrometer, equipped with a Broad Energy Germanium detector (BEGe). This detector has the advantage of covering the entire 0.3 to 3 MeV energy range with a good efficiency. The detector has been calibrated with a Monte Carlo probabilistic code.[9] The high resolution dual X-γ-spectrometry is performed[9] using a sophisticated software named AATAMI,[10] which accounts for the Breit-Wiegner X-ray energy distribution and the modeling of the shape of the background under the peaks. Table 2 summarizes the radioxenon minimum detectable activity concentration (MDC) calculated using their associated X-rays and γ-lines. It can be seen that the ^{131m}Xe MDC is improved respectively by a factor close to 10 if one considers a blank spectrum (empty cell) and close to 4 if one considers samples with very low ^{133}Xe activity concentration. In this case the sensitivity of ^{133m}Xe is only improved by a factor of 1.4. Table 2 also shows that ^{131m}Xe MDCs (resp. ^{133m}Xe) calculated from X-rays become quite equal and then larger than those calculated from γ-rays as soon as the activity of ^{133}Xe is greater than 500 mBq/m^3 (resp. about 40 mBq/m^3 in the case of ^{133m}Xe).

Mesoscale atmospheric transport modeling

A SPALAX™ system was set up at the CEA research centre in Bruyères-le-Châtel (France) in August 2003. Since then, it has been operating over 24-hour sampling time with a detection limit less than 1 mBq/m^3 for ^{133}Xe. Given the nuclear activity in Europe, a detectable amount of ^{133}Xe occurs in Bruyères-le-Châtel daily while the other isotopes ^{135}Xe, ^{133m}Xe, and ^{131m}Xe are detected only from time to time (Fig. 2). Between July 2003 and March 2006 (Fig. 3), about one hundred peaks with ^{133}Xe atmospheric volumetric activities ranging from ten or so to a few hundreds of mBq/m^3 stand out against a background activity level of some mBq/m^3. In order to determine the potential xenon sources, which may be situated up to some hundreds of kilometres away, a numerical study has been carried out.

Table 1. Factor of merit for the different radioxenon measured by X- and γ-spectrometry (nuclear data references)[7,8]

	^{133}Xe		^{135}Xe		^{131m}Xe		^{133m}Xe	
$T_{1/2}$, day	5.25		0.381		11.84		2.19	
E, keV	30.62–30.97 ($K_{\alpha1}$+$K_{\alpha2}$)	81	30.62–30.97 ($K_{\alpha1}$+$K_{\alpha2}$)	249.79	29.46–29.78 ($K_{\alpha1}$+$K_{\alpha2}$)	163.93	29.46–29.78 ($K_{\alpha1}$+$K_{\alpha2}$)	233.22
I	0.403	0.38	0.0402	0.902	0.442	0.0196	0.459	0.082
F	0.58	1.90	0.058	2.20	0.611	0.067	0.64	0.22

Table 2. Radioxenon MDC's (in mBq/m^3) calculated from both X- and gamma-rays from different spectra corresponding to an empty cell and samples containing increasing amounts of ^{133}Xe

MDC	Empty cell	Sample with increasing ^{133}Xe activity concentration, mBq/m^3					
		<0.11	12.1	46.3	225	575	1050
^{131m}Xe							
γ-ray, 164 keV	1.62	4.13	5.25	5.37	5.62	5.37	5.25
Kα X-ray	0.15	0.80	1.38	1.63	3.50	5.62	7.62
^{133m}Xe							
γ-ray, 233 keV	1.10	1.25	1.38	1.25	1.38	1.25	1.25
Kα X-ray	0.35	0.90	1.00	1.75	3.75	6.00	8.12
^{133}Xe							
γ-ray, 81 keV	0.11	0.18					
^{135}Xe							
γ-ray, 250 keV	0.18	0.25					

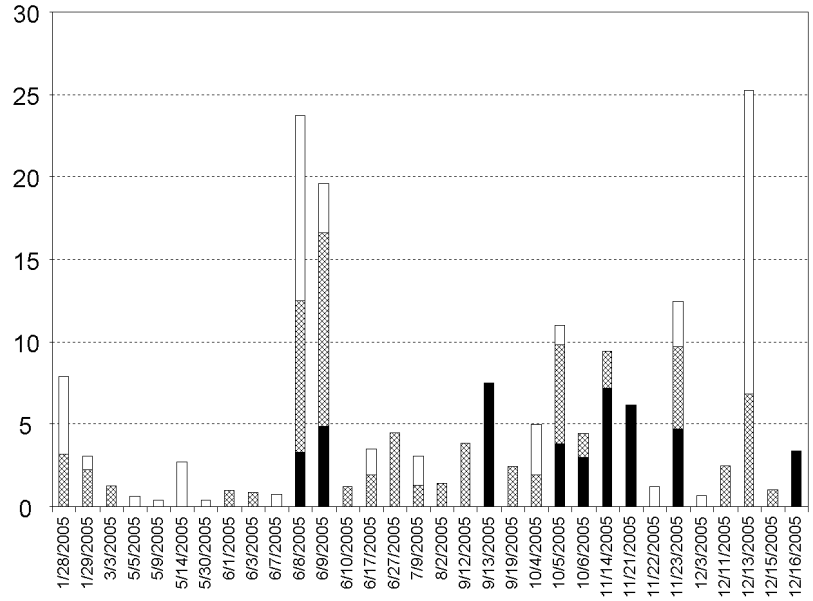

Fig. 2. Simultaneous occasional detections of isotopes (mBq/m^3): ^{131m}Xe (black), ^{133m}Xe (grey), ^{135}Xe (white). ^{133}Xe is given in Fig. 3

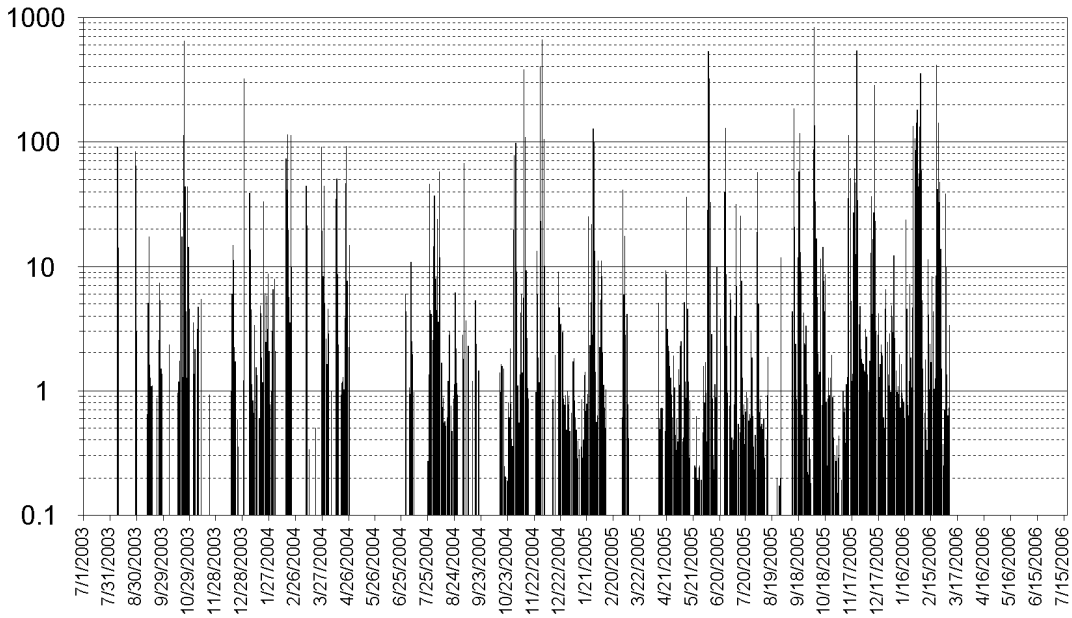

Fig. 3. Daily ^{133}Xe detections (in mBq/m^3 from 2003/07/01 to 2006/03/01)*Meteorological fields simulation*

The atmospheric modeling involves MM5 which uses a limited area modeling system to solve the non-hydrostatic compressible equations of the atmosphere dynamics on overlapping domains.[11]

Each MM5 simulation has the same characteristics. MM5 is run in the two-way nesting mode. Five grid lengths of 81 km, 27 km, 9 km, 3 km and 1 km are used to resolve successively finer scales. Roughly, the coarser resolution grid covers Western Europe. The five-domain vertical grid has 27 levels between the soil and an altitude of about 18,000 m (p_{top} = 100 hPa). The boundaries of the MM5 outer domain are issued every six hours with NCEP[12] analyses (horizontal resolution of 1°×1°). A 'moderate' relaxation towards the NCEP[12] analyses is applied for the five domains, except for the grid points in the PBL (Planetary Boundary Layer).

Backward transport simulation

The 3D diagnostic transport model FLEXPART[13] has been run in inverse mode to determine regions. To determine regions from which xenon may originate and to attribute sources to the detections. It is integrated backward in time and computes the trajectories of numerous particles representing air parcels with radioxenon source supposed releasing at the SPALAX™ detection site. A pre-processor has been developed in order to use the MM5 meteorological fields as input data in FLEXPART. A key parameter of the atmospheric dispersion is the boundary layer height also coming from MM5. The particles are emitted at each time step, from a box of 500 m×500 m, 20 m high, centred at 50 m above the ground level.

FLEXPART calculates the adjoint concentration field on a 3D grid similar to one of the MM5 grids, horizontally but not vertically (11 levels from the soil to 5,000 m). The concentrations are averaged on a time period of 600 s and stored in every 600 s for an air layer between 0 and 50 m. The simulations take account of ^{133}Xe radioactive decay ($\tau_{1/2} = 5.25$ day), while they assumed no significant dry, nor wet, deposition of xenon.

Inventory of potential sources

The region monitored by the Bruyères-le-Châtel SPALAX™ has a complex radioxenon background characteristic of an industrialized environment with a significant amount of nuclear facilities. The identified potential sources are located at the local, regional and mesoscale and they can be classified in three categories: nuclear power reactors and research reactors, industrial producers of radionuclides as well as medicine and research. Some examples of potential xenon sources belonging to each categories are the nuclear power plants (NPPs) in Western Europe; the commercial and research facilities operated in Fleurus (Belgium) to produce medical radioisotopes; and hospitals with nuclear medicine departments (especially in and around Paris). Due to extremely weak xenon releases, it is very unlikely that hospitals and research laboratories contribute significantly to the detections by the SPALAX™ in Bruyères-le-Châtel. On the other hand, more likely sources appear to be the European NPPs and the Belgian medical radioisotopes facility (Fig. 4).

Example analysis of a detection event

To illustrate the results, a particular detection event is analyzed because of both the complex meteorological situation and the multiplicity of the potentially implied xenon sources. A 24-hour average value of 33 mBq/m^3 in ^{133}Xe was measured in the air sampled by the SPALAX™ from 21 January 2004 at 10:00 UT to 22 January 2004 at 10:00 UT.

The wind field was simulated with MM5 and FLEXPART for the five days preceding the detection event. Figure 5 presents the ^{133}Xe backward activity concentration field, in the air layer from 0 to 50 m above the ground level, at six successive instants (from 22 Jan. 09:55 to 20 Jan. 12:05).

For each source, the backward ^{133}Xe activity concentration history is utilized to compute the magnitude of the release, potentially leading to the 33 mBq/m^3 in ^{133}Xe on 21 January 2004. Most of the nuclear power reactors seem to have a minor influence on the detection because there are situated quite far from the SPALAX™ and they usually do not emit such high amounts of ^{133}Xe. The case of Nogent-sur-Seine reactor is different because this NPP is quite close to Bruyères-le-Châtel and the meteorological situation was partly propitious to a potential detection owing to an easter wind and a stable atmosphere. Though located farther from the SPALAX™, the medical radioisotopes production facility in Fleurus (Belgium) also constitutes a relevant source as the meteorological flux was from the north-east for a part of the considered period. In both cases, the calculated required ^{133}Xe releases, leading to the detection on 21 January, are consistent with the orders of magnitude of realistic releases in normal operation from Nogent-sur-Seine reactor and/or Fleurus facilities.

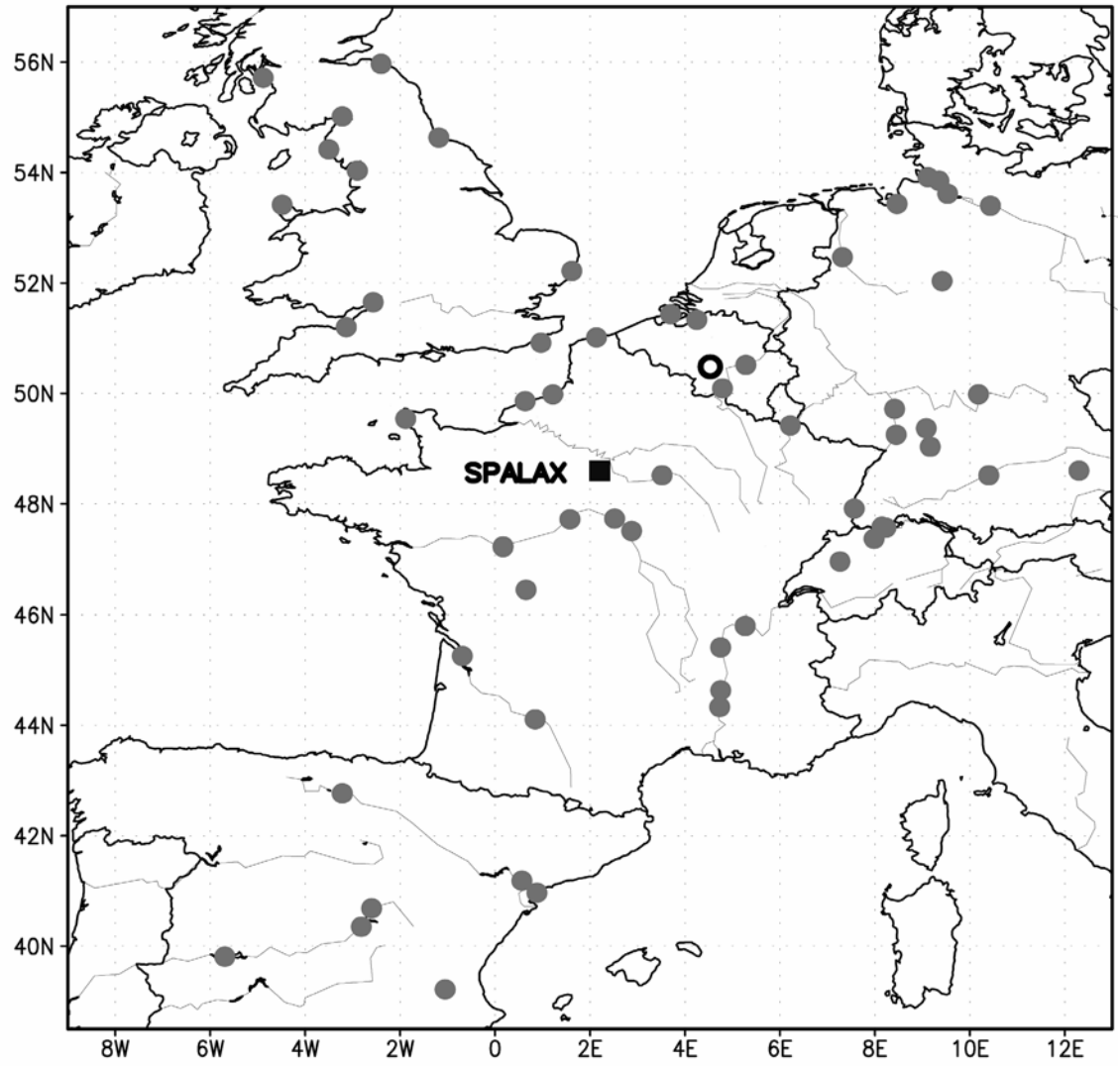

Fig. 4. Location of the SPALAX™ detector and major potential sources identified in the calculation domain (● European NPPs; ○ medical isotope facility)

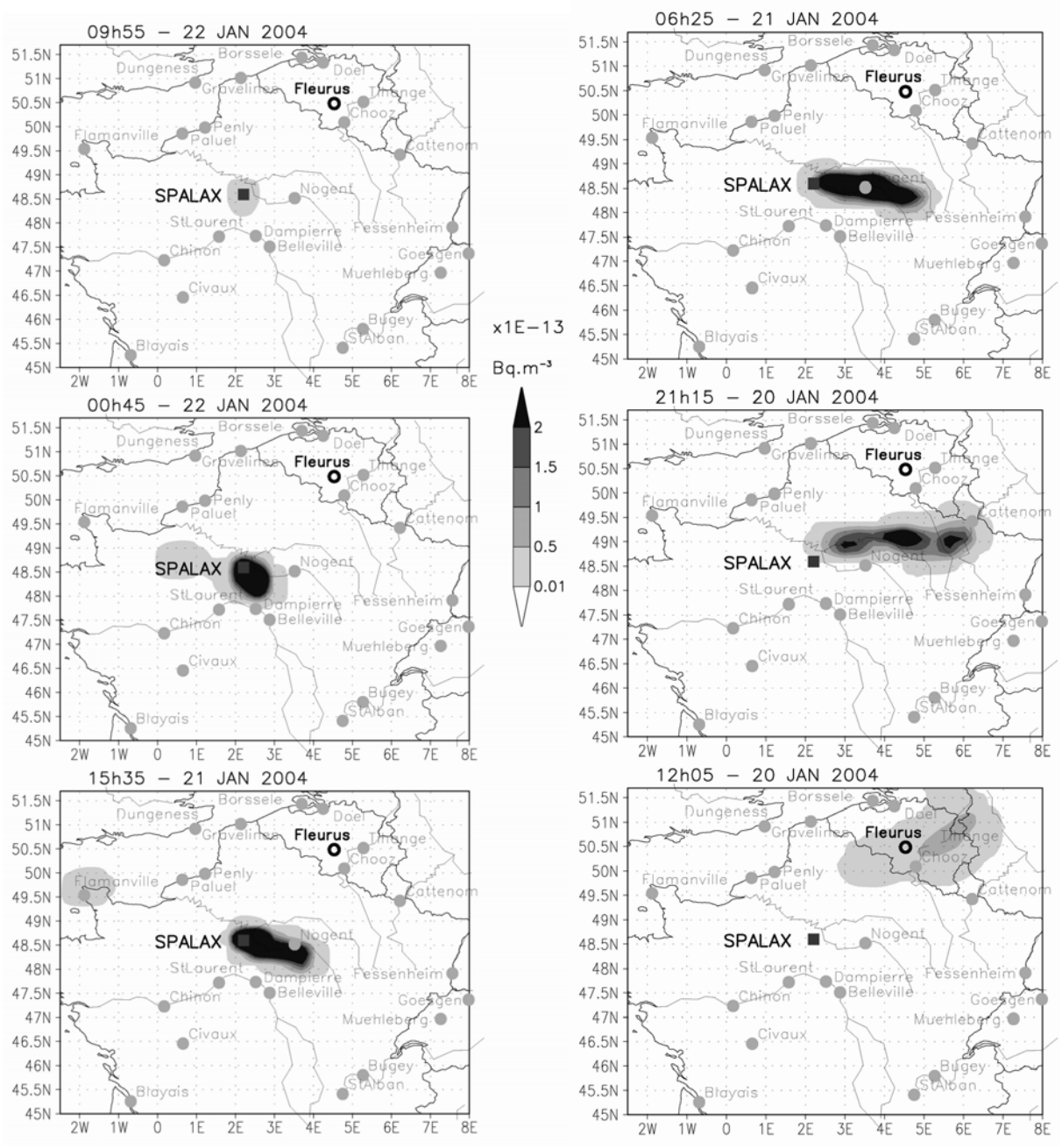

Fig. 5. FLEXPART results. Backward activity concentration of ^{133}Xe from 22 January 2004 at 09:55 UT to 20 January 2004 at 12:05 UT

Conclusions

In order to fulfil the requirements and to support the CTBTO for verifying the Compliance with the treaty, our contribution led us to develop the following set of tools: (1) an automated fieldable radioxenon collection plus detection system, (2) a sensitive and reliable radioxenon analysis at the laboratory and (3) a mesoscale atmospheric transport modeling methodology.

In this paper an atmospheric transport modeling system (MM5 weather prediction suite and FLEXPART Lagrangian dispersion model) was used to analyze and explain the radioxenon events detected by the SPALAX™ gas system located at CEA Bruyères-le-Châtel. Some samples were measured more precisely in the laboratory using dual X-γ-spectrometry after transfer of the xenon from the archive bottle to a counting cell. In the context of the environmental monitoring and CTBT compliance verification, the motivation is to

determine the radioxenon sources by computation of back-trajectories from the detector in Bruyères-le-Châtel. While the estimation of a potential source is expected to be quite simple in regions devoid of nuclear industry, it happens that the SPALAX™ is under the influence of multiple and complex sources located in Western Europe.

For the ^{133}Xe detection on 21 January 2004, as for almost all detection events, many sources have a significant emission efficiency and are potentially implied. The calculated source terms required to justify the radioxenon detection must be compared with the inventoried sources in order to determine the most likely source(s). In the paper, it is demonstrated that the 'local' sources (like the hospitals, research laboratories…) in the Paris region are quite unlikely to induce significant radioxenon detections in Bruyères-le-Châtel. On the contrary, the 'regional' nuclear power reactors, and the far away, the medical radioisotopes production facility in Fleurus (Belgium) are probably involved in the ^{133}Xe detections. In order to reach a unique solution it will be necessary to analyze others parameters, in particular the isotopic composition of the source terms.

References

1. Comprehensive Nuclear-Test-Ban-Treaty (CTBT), Provisional Technical Secretariat, Test on the Establishment of a Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organisation, 1996.
2. J. P. FONTAINE, F. POINTURIER, X. BLANCHARD, T. TAFFARY, J. Environ. Radioact., 72 (2004) 129.
3. T. W. BOWYER, Nuclear Instruments and Methods in Physics Research, Encyclopedia of Environmental Analysis and Remediation, R. A. MEYERS (Ed.), John Wiley, 1998.
4. Y. FINKELSTEIN, Kerntechnik, 66 (2001) 229.
5. KALINOWSKI, K. UNGAR, T. W. BOWYER, Appl. Radiation Isotopes, to be published.
6. SCHOENGOL, M. E. DEMARRE, E. M. KIRKWOOD, Radiological Effluence Released from US Continental Tests through 1961 to 1992 US DOE DOE/NV-317 (Rev. 1) UC-702, Las Vegas, 1996.
7. T. SIISKONEN, H. TOIVONEN, Radiat. Phys. Chem., 69 (2004) 23.
8. LBNL Isotopes Project- LUNDS Universtet Version 2.1 http://ie.lbl.gov./toi/
9. G. LE PETIT, C. JUTIER, P. GROSS, V. GREINER, Appl. Radiation Isotopes, 64 (2006) 1307.
10. CTBTO Preparatory Commission, User Manual of Radionuclide Analysis and Evaluation Software, Aatami version 3.04, Evaluation Section, Vienna, 2002.
11. MM5, PSU/NCAR Mesoscale Modelling System, MM5. Tutorial Class Notes and User's Guide. Version 3 (release 3–6), 2003.
12. NCEP, National Centers for Environmental Prediction, http://www.ncep.noaa.gov
13. A. STOHL, M. HITTENBERGE, G. WOTAWA, Atmospheric Environment, 32 (1998) 4245.