ANALYSIS OF ATMOSPHERIC RADIOXENON ACTIVITIES MEASURED BY A RADIONUCLIDE GAS STATION LOCATED IN FRANCE: SIMULATION OF THE ATMOSPHERIC TRANSPORT WITH A MESOSCALE MODELLING SYSTEM

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CONTEXT AND OBJECTIVES

The detection and measurement of radionuclides released in the frame of nuclear activities is a major task addressing various issues like the monitoring of the environment for regulatory requirements, or the verification of the compliance with the Comprehensive nuclear Test Ban Treaty (CTBT).

In the late 1990s, the French Atomic Energy Commission (CEA) developed a high sensitive technology, capable of performing radioactive noble gas measurements. This apparatus, called SPALAX for *Système de Prélèvement d'air Automatique en Ligne avec l'Analyse des radio-Xénons*, is described by *Fontaine et al.* (2004). The SPALAX automatically extracts, purifies and concentrates gaseous xenon from the atmospheric air; it measures the volumetric activity of four radioxenon isotopes ^{131m}Xe, ^{133m}Xe, ^{133}Xe, ^{135}Xe using high resolution γ spectrometry.

The radioxenon isotopes are fission products chronically released from many kinds of nuclear installations such as nuclear power reactors, research reactors, hospitals and medical isotopes production facilities. The magnitudes and the proportions of the four radioactive isotopes may be useful to discern the various nuclear industry xenon emissions.

A SPALAX station was set up at the CEA research centre in Bruyères-le-Châtel (located 40 km to the south of Paris) in August 2003. Since then, it has been operated with a 24 hour-sampling time. With the very low detection limit afforded by the SPALAX technology (e.g. less than 1 mBq.m^{-3} in ^{133}Xe) and the nuclear European environment, 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. The four isotopes were seldom observed simultaneously.

Between August 2003 and August 2005, *ca.* eighty 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}. These values are far below any levels of health concern.

In order to explain the radioxenon detections in Bruyères-le-Châtel, a numerical study has been carried out. The purpose is the determination of the potential xenon sources, which may be local or situated up to some hundreds of kilometres, generating the SPALAX observations. Thus, the atmospheric transport modelling requires a mesoscale approach which is described in the paper. We report successively on the meteorological fields and backward transport simulations performed for each event of xenon detection. The use of the mesoscale modelling is illustrated by the analysis of a quite elevated ^{133}Xe measurement in Bruyères-le-Châtel. Finally, the future prospects of the numerical approach are drawn.

METEOROLOGICAL FIELDS SIMULATION

The atmospheric transport modelling involves the MM5 suite developed at Pennsylvania State University and National Centre for Atmospheric Research. MM5 is a limited area modelling system used to solve the non-hydrostatic compressible equations of the atmosphere dynamics. The calculations are run on nested domains with horizontal resolutions from 1 km to 100 km.

The 'MM5 code' itself is the last part of a modelling system constituted of modules dedicated to the definition of the calculations domains (location, size, resolution, etc.), the interpolation of the terrain elevation, land-use and soil categories on the surface grids, and the interpolation

of global meteorological analyses on the 3D grids as MM5 requires an initial condition, and lateral and surface boundary conditions for the entire simulation. MM5 weather prediction is performed by a terrain-following, sigma-coordinate model including many advanced physical parameterisations related to the physics of precipitations, the planetary boundary layer (PBL) and surface layer processes, the radiation, etc. (*MM5*, 2003).
Each MM5 simulation carried out to analyse the radioxenon detections in Bruyères-le-Châtel 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 the Western Europe. The finer resolution grid zooms in on Paris and vicinity (Figure 1). 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 MM5 outer domain are issued every six hours with NCEP analyses (horizontal resolution of 1° x 1°). MM5 physical parametrisations are the standard and robust ones. A 'moderate' relaxation towards the NCEP analyses is applied for the five domains, excepted for the grid points in the PBL. Indeed, it has been shown that the 'analysis nudging' notably improves the flow prediction in the free troposphere, and also in the boundary layer.
The meteorological fields have to be as accurate as possible to warrant the precision of the dispersion calculations. The quality of the wind fields is assessed by comparing the results of MM5 simulations with the observations at Bruyères-le-Châtel meteorological mast and at Météo-France meteorological stations. In most cases, the agreement is excellent between the calculations and the measurements of the wind modulus and wind direction. The high MM5 space and time resolution is capable of reproducing the quick variations in the intensity and direction of the wind.

BACKWARD TRANSPORT SIMULATIONS

The 3D diagnostic transport model FLEXPART has been run in inverse mode to support the interpretation of the measurement data gathered in Bruyères-le-Châtel.
FLEXPART is a Lagrangian code developed at Munich University by *Stohl et al.* (1998). It is integrated backward in time and computes the trajectories of numerous particles representing air parcels with radioxenon released by the SPALAX. A pre-processor has been elaborated in order to use the MM5 meteorological fields as input data in FLEXPART. A major advantage of the model is to deal with nested domains. The velocity of the particles is characterised by a mean component resulting from MM5, and a stochastic component calculated by FLEXPART according to *Thomson* (1987). 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 x 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. The fields represented later on are average activity concentrations in the air layer between 0 and 50 m. The simulations take account of ^{133}Xe radioactive decay ($\tau_{1/2}$ = 5.25 days), while there is no significant dry, nor wet, deposition of xenon.
The backward transport calculations aim at determining regions from which radioxenon may originate and attributing sources to the measurements by the SPALAX in Bruyères-le-Châtel. The principle is as follows: for each detection event, a backward source located at the detector position emits continuously the activity q (in Bq) during the sampling period (24 hours). One could notice that usually $q = q_{unit}$ = 1 Bq. The tracer is advected and dispersed by FLEXPART and the time profiles of the backward activity concentration c^* (in Bq.m^{-3}) are recorded for the identified potential sources. For each given source, the magnitude of the release Q (in Bq), of duration T_s (in s), required to obtain the 24 h-average activity concentration $\bar{c}$ at the detector, is evaluated by the formula (1) below. The minimum release Q_{min} is 'instantaneous' (in fact,

of duration t_s = 600 s) and corresponds to the maximum value c^*_{max} of the backward activity concentration recorded for the source. It is given by formula (2).

$$Q(T_s) = \frac{q \,.\, \bar{c}}{\frac{1}{T_s} \int_{T_s} c^*(t)\, dt} \qquad (1) \qquad\qquad Q_{\min} = \frac{q \,.\, \bar{c}}{c^*_{\max}} \qquad (2)$$

INVENTORY OF POTENTIAL SOURCES

It is known that in normal operation, various nuclear installations chronically or sporadically release small amounts of radioxenon in the atmosphere. In order to supplement the numerical study and to explain the xenon detections by the SPALAX in Bruyères-le-Châtel, it has been decided to proceed to the inventory of the potential radioxenon sources in the western part of Europe. Although this work is still in progress, some results are reported hereafter.

The region monitored by 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: the nuclear power reactors & research reactors; the producers of radionuclides used in the industry, the medicine and the research; finally, the radionuclides users for medicine and research. Some examples of potential xenon sources belonging to each categories are the Nuclear Power Plants (NPPs) in west 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 installation (Figure 2). The search for xenon potential sources by means of the ATM will confirm this estimation.

Figure 1: The five nested grids D01 to D05 of MM5 simulations. The dimensions are respectively 3’321 km x 2’835 km,
1’404 km x 1’323 km, 522 km x 522 km,
210 km x 210 km, and 64 km x 79 km.

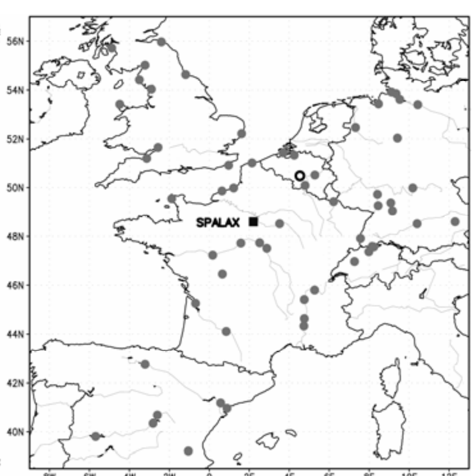

Figure 2: Location of the SPALAX detector and major potential sources identified in the calculation domain (circle: European NPPs; ring: medical isotopes facility).

EXAMPLE OF ANALYSIS OF A DETECTION EVENT

Almost all radioxenon detection events in Bruyères-le-Châtel have been numerically studied. To illustrate the results, this paper focuses on a particular detection considered as interesting because of both the complex meteorological situation and the multiplicity of the potentially implied xenon sources. A 24h-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.

Making use of our atmospheric transport modelling, the mesoscale wind field was simulated with MM5 for the five days preceding the detection event. FLEXPART was used to compute the back-trajectories of particles emitted continuously from the SPALAX location during the considered 24h-sampling period. Derived from the time and space distribution of the particles, Figure 3 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).

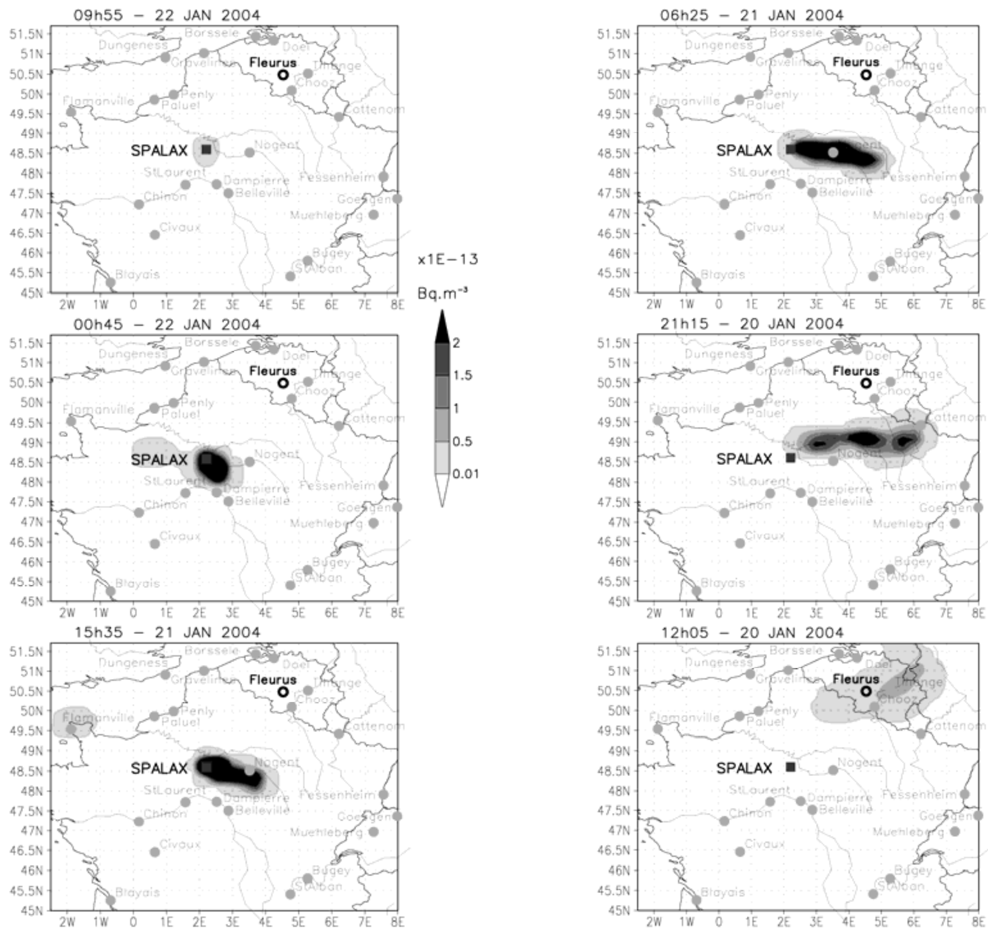

*Figure 3: FLEXPART results. Backward activity concentration of ^{133}Xe
from 22 January 2004 at 09:55 UT to 20 January 2004 at 12:05 UT.*

From a meteorological point of view, the three-day time period in Figure 3 was characterized by reversals of the wind. On 20 January, the general flux was from the north-east and rotated to the east in the evening. On 21 January, the wind first blew from the east, and progressively reversed to the east-south-east. From 21 January evening to the end of the SPALAX detection (22 January at 10:00 UT), the wind was weak with a very variable direction. Furthermore, the mixing height (computed by the PBL scheme in MM5) was low during most of the episode denoting a very stable atmosphere, especially on 21 January. This description illustrates the complex meteorological situation associated with the studied detection.

Figure 3 shows that many sources could be involved in the ^{133}Xe measurement on 21 January 2004. Indeed, the backward plume passes over the medical isotopes installation in Fleurus, NPPs located in Belgium and in the north-east (and even in the west) part of France, and also Paris region with its hospitals and nuclear research laboratories. Recorded for all the potential sources, the time profiles of the backward ^{133}Xe activity concentration inform on the efficiency and likelihood of the sources. The temporal series are used to get the magnitudes of the releases leading to the 33 mBq.m^{-3} in ^{133}Xe on 21 January 2004. Even located close to the SPALAX detector, the Parisian local sources should exhibit elevated emissions, unrealistic in relation to these facilities. Most of the nuclear power reactors seem to have a minor influence on the detection as, situated quite far from the SPALAX, they should have unlikely high ^{133}Xe emissions. The case of Nogent-sur-Seine reactor is different because it is one of the closest NPPs and the meteorological situation was partly propitious to a potential detection owing to an east flux 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.

CONCLUSIONS

This paper describes our atmospheric transport modelling system, based on the MM5 suite for the prediction of the meteorological fields and FLEXPART Lagrangian dispersion model. It was elaborated to analyse and explain the radioxenon detections obtained at the SPALAX gas station located in Bruyères-le-Châtel CEA research centre. In the context of the environmental monitoring and compliance verification of the CTBT, the motivation is the spotting of the space and time location of the xenon sources.

The paper comments on the particular ^{133}Xe detection event on 21 January 2004, taken as an example of xenon sources attribution 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 our SPALAX is under the influence of multiple and complex sources located in Western Europe. Thus, it is essential to proceed to a radioxenon sources inventory as comprehensive as possible.

The atmospheric transport modelling indicates the regions from which the radionuclides may originate. For the considered ^{133}Xe detection as for almost all detection events, many sources have a significant emission efficiency and are potentially implied. The calculated source terms required to get the detection recorded by the SPALAX must be compared with the inventoried sources in order to suppress unrealistic estimations in terms of contributing facilities.

Applying this method, it has been inferred that the so-called ‘local’ sources (like the hospitals, research laboratories…) in Paris region are quite unlikely to induce the significant radioxenon detections in Bruyères-le-Châtel. On the contrary, the ‘regional’ nuclear power reactors, and far away, a medical radioisotopes production installation in Fleurus (Belgium) are probably involved in the ^{133}Xe detections. Further work together with contact with the operators of the facilities are required before concluding.

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