CONTRIBUTIONS OF XENON RELEASES IN THE ATMOSPHERE FROM RADIONUCLIDE PRODUCTION FACILITIES AND NUCLEAR POWER PLANTS TO THE DETECTION OF 133XE BY SPALAX SYSTEMS IN WESTERN EUROPE

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INTRODUCTION

The Comprehensive nuclear Test Ban Treaty (CTBT) bans nuclear explosions in all environments. In the frame of CTBT monitoring, the French Atomic Energy Commission (CEA) has developed a system, called SPALAX (French acronym for "Système de Prélèvement d'air Automatique en Ligne avec l'Analyse des radio-Xénons"), to automatically measure the activity concentrations of four radioactive isotopes of xenon, ^{131m}Xe, ^{133m}Xe, ^{133}Xe and ^{135}Xe, in the atmosphere [*Fontaine et al., 2004*]. Among these isotopes, ^{133}Xe is the most frequently detected and in highest concentrations. Measurements of ^{133}Xe by three SPALAX systems (figure 1) operated in Bruyères-le-Châtel (called DIF in the following, France), Schauinsland (called Freiburg in the following, Germany) [*Schlosser, 2006]* and temporarily in Marseilles (France), show a baseline of a few mBq.m^{-3} and peaks from a few tens to a few hundreds of mBq.m^{-3}. In order to explain these radioxenon detections, a numerical study of atmospheric dispersion has been carried out.

METHOD

Our Atmospheric Transport Modelling system (ATM) is based on the PSU/NCAR Mesoscale Model (MM5) and the particle dispersion model FLEXPART. MM5 is a limited area, nonhydrostatic, terrain following and sigma-coordinate meteorological field model [*MM5, 2003*]. In this study, each MM5 simulation is run in the two-way nesting mode and a relaxation towards the GFS analysis (6 hours, 1° × 1°) is applied for the domains. Standard physical parameters have been used. FLEXPART is a Lagrangian Particle Dispersion model, widely used to calculate the transport of non-reactive pollutants in the atmosphere [*Stohl et al. 1998*]. A version is available to handle the nested outputs of the mesoscale MM5 model.

POTENTIAL SOURCES OF ATMOSPHERIC XENON-133

Western Europe has a complex radioxenon background. The identified potential sources can be classified in three categories:

- The Nuclear Power Plants (NPPs). In this study we have assumed that each NPP produces a continuous release of ^{133}Xe in the atmosphere. As a first approximation, we have considered that the preponderant releases are the routine releases excluding unit outages. In this work, they have been represented by a continuous release of a few GBq / day / reactor (1 GBq = 1E9 Becquerel).
- The medical radioisotope production facilities. In the northern-hemisphere, there are two major plants located at Fleurus (Belgium) and at the Chalk River Laboratory, close to Ottawa (Canada). The installations report daily releases with activity ranging from a few TBq (1 TBq = 1E12 Becquerel) to a few tens of TBq for Fleurus and daily releases of a few tens of TBq for Chalk River.
- The hospitals with nuclear medicine departments and research laboratories. Due to their extremely weak xenon releases, we have assumed that these installations do not contribute significantly to the SPALAX detections.

OCTOBER-NOVEMBER 2004 DETECTIONS

With these assumptions, we have simulated the atmospheric transport of xenon emitted by the Fleurus facility and by NPPs on the scale of Western Europe (figure 1). Obtaining the spatial and temporal distribution of ^{133}Xe, we have evaluated the concentration activities on the three SPALAX. Simulations have been carried out during the period from October 25, 2004 to November 16, 2004. This particular period has been chosen since continuous measurements and significant peaks have been measured on the three SPALAX systems.

Meteorological field simulation

We defined two nested domains with a 81 km horizontal resolution, close to the GFS analyses resolution, for the coarse domain, and a resolution of 27 km for the inner domain. Figure 2 shows the geographical extent of the two domains.

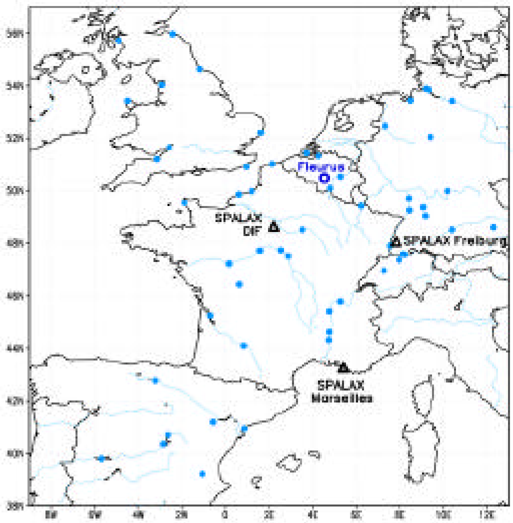

Fig. 1; Location of the SPALAX detectors and major potential sources (circle: NPPs; ring: medical isotope production plant).

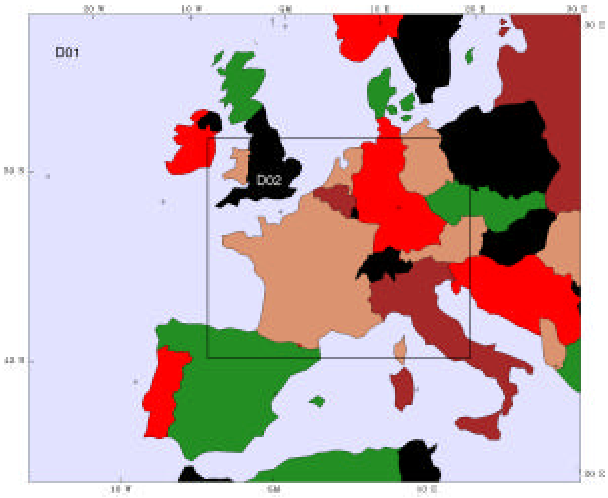

Fig. 2; Domain setup: D01 (resolution of 81 km) and D02 (resolution of 27 km).

For the considered period, we have validated the MM5 simulation by comparing the results with meteorological observations. For example, figure 3 shows that the wind direction calculated at 10 m above ground level is in good agreement with surface observations of the Trappes radiosounding station (close to the SPALAX DIF).

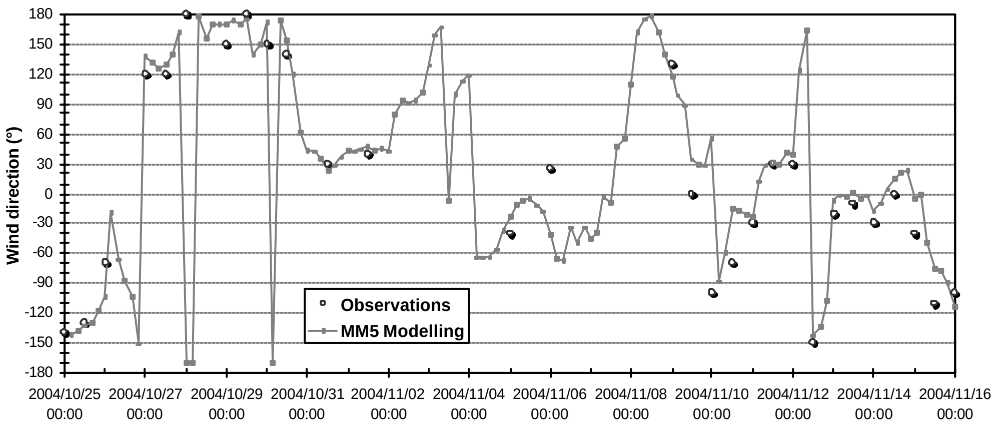

Fig. 3; Wind direction from October 25 to November 16, 2004 simulated using MM5 and observations from radiosoundings at Trappes (France).

Transport and dispersion results

For each considered SPALAX, figure 4 shows the comparison of the 24h-averaged ^{133}Xe activity concentrations (log scale) measured (dashed line) and calculated by our ATM (grey pattern). The peak heights are reproduced correctly using relatively simple assumptions about xenon emissions. The time synchronization is very good to a first order approximation. The releases from the western European NPPs generate a baseline of the order of one to about ten mBq.m^{-3} and the releases from radionuclides production facilities lead to major peaks of the order of tens to a few hundreds mBq.m^{-3}. Thus, radioxenon emissions from Fleurus and the western European NPPs are necessary and sufficient to account for the ^{133}Xe levels detected by the SPALAX instruments at DIF, Freiburg and Marseilles.

The main contributors to the ^{133}Xe measurements of the SPALAX detectors, but also throughout western Europe, are thus probably identified.

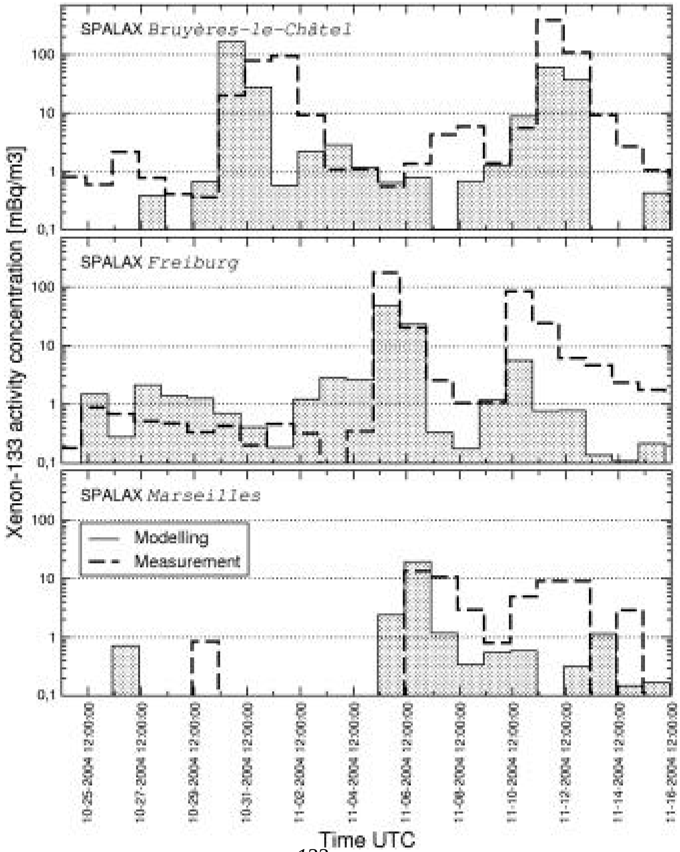

Fig. 4; 24h-averaged ^{133}Xe activity concentrations in mBq.m^{-3} for the three SPALAX: simulations (grey pattern); measurements (dashed line).

JANUARY 17, 2006 DETECTION EVENT AT BRUYÈRES-LE-CHÂTEL

As shown before, most of the significant SPALAX detections in Western Europe are related to releases from the Fleurus plant. However, some detections can not be fully explained with such releases or routine operation from NPPs. In these cases, other potential sources have to be considered, such as the Chalk River radioisotope production plant (Canada) since the facility produces daily significant ^{133}Xe releases. In this part, we discuss the analysis of one of these particular detections which took place on January 17, 2006 at Bruyères-le-Châtel.

Meteorological field simulation

According to the method described above, two nested domains with 81 km and 27 km horizontal resolution respectively were defined to calculate the wind field from January 3 to 21, 2006. The coarse domain includes the Chalk River plant and a part of Europe. The finer domain includes the SPALAX DIF and the major western European identified sources (Fig. 5).

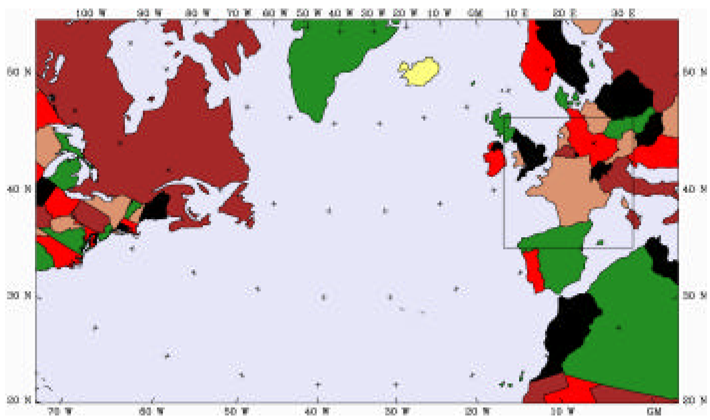

Fig. 5; Domain setup: D01 (81km res.), D02 (27km res.).

For the considered period, MM5 results were compared to surface observations. Figure 6 shows that the wind direction calculated at 10 m fits very well the observations made at the Paris/Orly airport METAR station (close to the SPALAX DIF system). During the collection time of the detector (Jan. 17, 2006 at 10 a.m. to Jan. 18, 2006 at 10 a.m.) the wind was from the south and rotated to the north-west before coming back to the west / south-west. It is important to notice that Fleurus facility releases reach directly the SPALAX DIF when the wind is from north-east. Such a situation is not observed during the few days before sampling.

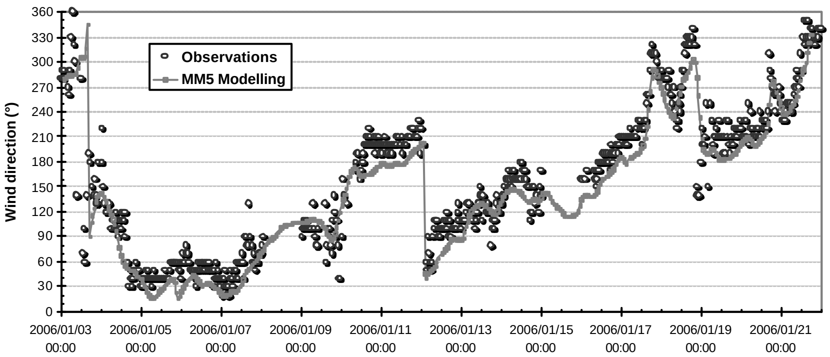

Fig. 6; Wind direction from January 03 to 22, 2006 of MM5 simulations and observations taken from METAR network at Orly (France).

Transport and dispersion results

We have simulated the atmospheric transport of xenon emitted by the main European sources (Fleurus and NPPs) and by the Chalk River facility. No accidental release from NPPs has been declared for this period. Figure 7 shows that releases from European sources lead to weak simulated peaks for all the period. Moreover, due to the wind direction, releases from Fleurus lead to non-zero signal only for January 12 and 13. The NPPs releases generate a baseline of the order of a few $mBq.m^{-3}$, which decreases at the end of the period.

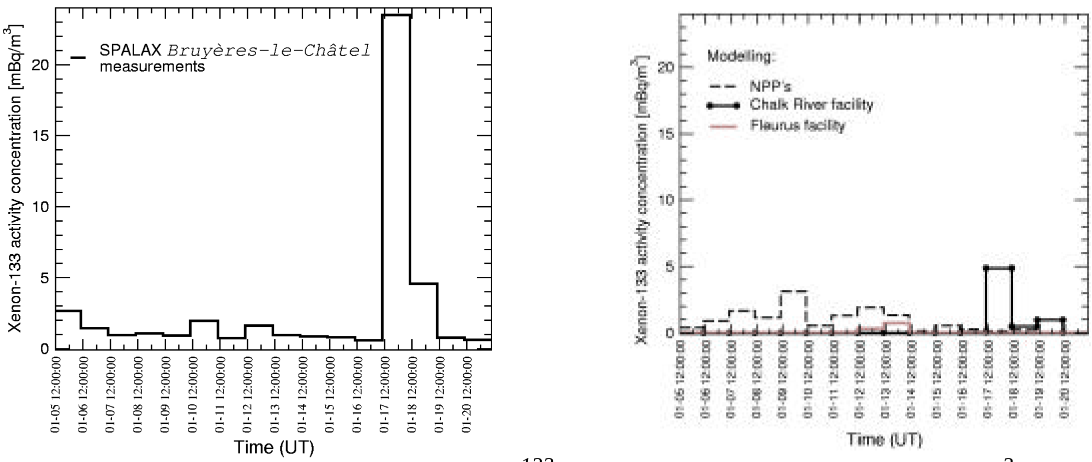

Fig. 7; Comparison of the 24h-averaged ^{133}Xe activity concentrations ($mBq.m^{-3}$) at the SPALAX DIF: measurements (left) and simulations (right).

Thus, these sources do not appear to contribute to the January 17, 2006 peak. On the other hand, even after a long range transport (about 6000 km), Chalk River releases lead to a significant calculated signal around Jan. 17. Figure 8 shows the 24h-averaged ^{133}Xe activity concentration ($mBq.m^{-3}$) calculated at the scale of France, assuming only releases from Fleurus (left figure) and only releases from Chalk River (right figure). It clearly appears that Chalk River could be the major contributor to this SPALAX detection.

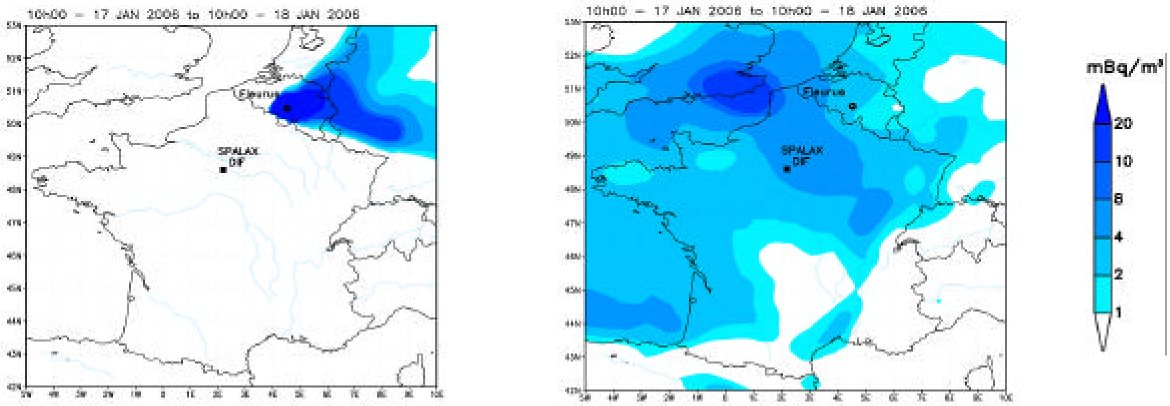

Fig. 8; 24h-averaged ^{133}Xe activity concentration (mBq.m^{-3}) simulated around the SPALAX DIF site from January 17, 2006 at 10 a.m. to January 18, 2006 at 10 a.m. Left: contribution of Fleurus releases. Right: contribution of Chalk River releases.

CONCLUSION

Considering average realistic emissions from Fleurus (Belgium) medical radioisotope production facility and West-European nuclear power plants (NPPs), atmospheric transport modelling (ATM) was performed to simulate the activity concentration of ^{133}Xe in Western Europe. The ATM includes MM5 and FLEXPART. The simulations have shown that, to a first-order approximation, calculated signals are in good agreement with measurements of the three SPALAX systems. We have pointed out that the releases from the western European NPPs generate a baseline of the order of one to about ten mBq.m^{-3} and the releases from radionuclides production facilities lead to major peaks of the order of tens to a few hundreds mBq.m^{-3}. Then, the main contributors to the ^{133}Xe measurements of the SPALAX detectors, but also throughout Western Europe, have been probably identified. However, some detections can not be fully explained with above mentioned releases. In these cases, other potential sources have to be considered, such as the Chalk River radioisotope production plant (Canada). Calculations including this source have shown that the contribution Chalk River plant to the measurements in Western Europe could be significant.

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