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Measurement and modelling of radioxenon plumes in the Ottawa Valley

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ABSTRACT

Since 2001 a real-time radiation monitoring network of Canadian nuclear facilities and major population centres has been implemented for response to nuclear incidents including a possible terrorist attack. Unshielded NaI(Tl) spectroscopic detectors are employed to measure gamma radiation from airborne radioactivity and radioactivity deposited on the ground. These detectors are composed of a standard $3'' \times 3''$ cylindrical NaI(Tl) spectrometers with data storage and integrated telemetry. Some of the detectors have been deployed in the Ottawa Valley near Chalk River Laboratories and Ottawa, which has a complex radioxenon environment due to the proximity of nuclear power reactors, and medical isotope facilities. Although not a health threat, these releases have provided an opportunity for the Canadian Meteorological Centre and the Commissariat à l'Energie Atomique to validate their meteorological models. The meteorological models of the two organizations are in good agreement on the origin and the source terms of these releases.

1. Introduction

The Ottawa Valley provides a unique location to test meteorological models using a fixed point NaI(Tl) spectrometer network in addition to the Système de Prélèvement d'air Automatique en Ligne avec l'Analyse des radioXénons (SPALAX) equipment. The spectrometer network was designed for response to nuclear incidents and the SPALAX for the compliance verification of the comprehensive nuclear-test-ban treaty. Both systems are effective tools for environmental monitoring for radioactivity, specifically airborne radioxenon and complement each other in terms of temporal resolution and activity concentration accuracy.

Ottawa has multiple radioxenon sources within a radius of a few hundred kilometers including hospitals and a medical isotope facility within city limits. A better understanding of the relative importance of these sources and the dispersion of radioxenon through the environment has been obtained through this work, providing further insight into the radioxenon background in the world.

Health Canada operates a network of identical fixed point NaI(Tl) spectroscopic detectors which are pointed skyward to measure dose from radioactive plumes (Ungar et al., 2003) and materials deposited on the ground. This network has been deployed as part of an emergency preparedness and counter terrorism initiative. These detectors are deployed in three types of locations:

(1) sub-networks of NaI(Tl) detectors around Canadian nuclear facilities,
(2) single NaI(Tl) detectors in major Canadian population centres,
(3) one to three NaI(Tl) detectors in population centres near nuclear-powered vessel mooring sites.

The purpose of this network is to ensure the health and safety of Canadians through the continual monitoring of radiation dose and to report emission levels in the case of a nuclear emergency. It is integrated into Health Canada's planned response in the event of a nuclear emergency.

In this study we focus on the sub-network of NaI(Tl) detectors around the Chalk River Laboratory nuclear facility, in the Ottawa Valley. Fig. 1 shows the detector locations.

Health Canada also operates radionuclide detection systems in Ottawa in support of compliance verification of the CTBT. The compliance regime once the treaty is in force uses four technologies: hydro-acoustic, infrasound, seismic, and radionuclide monitoring (Sullivan, 1998). The last technology is the only one

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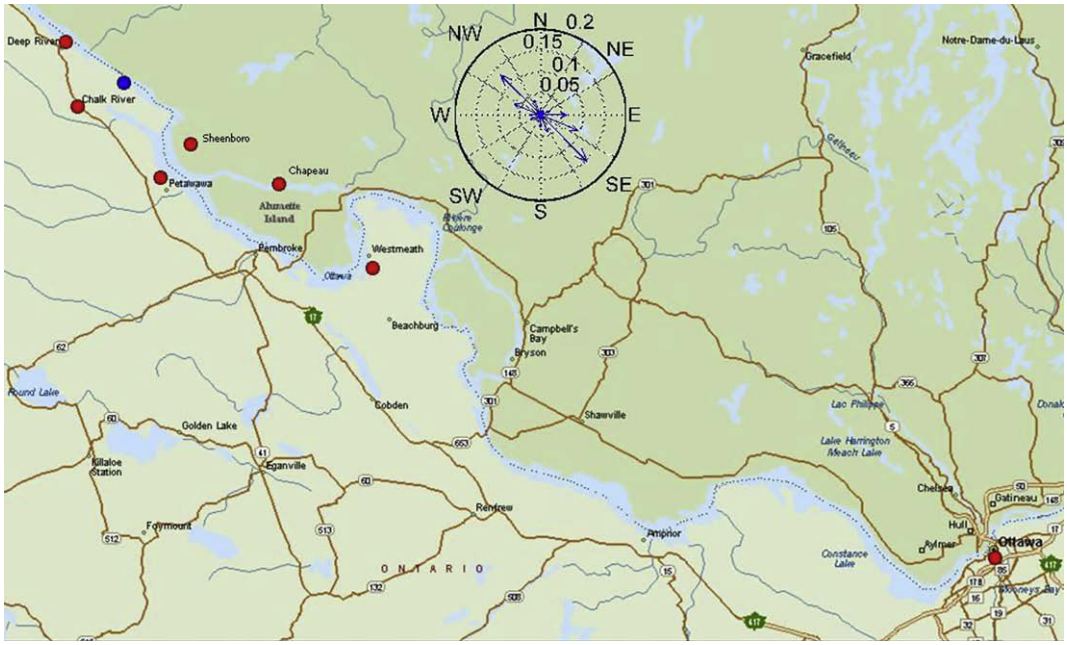

Fig. 1. A map of the Ottawa Valley, showing the NaI(Tl) monitoring network. Red dots indicate locations of the detectors (moving south-eastward) in Deep River, Chalk River Labratories (blue), town of Chalk River, Sheenboro, Petawawa, Chapeau, Westmeath, and Ottawa. A wind rose indicating the direction the wind travels at Chalk River Laboratories is also indicated (produced from data in Niemi et al., 2001). The arrow points in the direction towards which the wind is blowing. For example the wind travels from the west (easterly) 5% of the time.

which can unambiguously discriminate a nuclear test from other events and includes both radioactive particulate and radioactive noble gas monitoring. Upon entry into force, 40 noble gas monitoring stations are planned to verify the treaty. Currently, the Provisional Technical Secretariat of the CTBT Organization Preparatory Commission is conducting an International Noble Gas Experiment (INGE) consisting of 19 stations including Ottawa. Ottawa is not an official CTBT site but serves as a useful test-bed station.

For noble gas monitoring Canadian CTBT stations are equipped with SPALAX technology, designed and developed at Département Analyse, Surveillance, Environnement (DASE) laboratories of the French Atomic Energy Commission. The Ottawa instrument is co-located with one of the NaI(Tl) detectors of the Ottawa Valley sub-network. The SPALAX measures the activity concentrations of ^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe by separating and concentrating gaseous elemental xenon from the atmosphere and then measuring the gamma rays emitted via high resolution spectrometry. The SPALAX is more thoroughly described elsewhere (Fontaine et al., 2004; International Patent 1999; Auer et al., 2004).

The ratios and magnitudes of these four isotopes, isomer combinations vary depending on the source; for example, nuclear explosions, fuel processing, electricity generation, medical isotope production, medical isotope use, or other human activity. A study of the SPALAX measurements in Tahiti and in Ottawa is given by Stocki et al. (2005). The half-lives of these four species are given in Table 1.

The SPALAX and the Ottawa Valley NaI(Tl) sub-networks provide powerful tools for the detection and tracking of radioxenon within the Ottawa Valley. These data were employed for the testing of meteorological models as radioactive plumes propagated through the complex valley terrain. By comparing these measurements with different atmospheric transport models from two institutions, insight can be gained in the physics and techniques used. These comparisons lend confidence to the meteorological models employed.

Section 2 of this paper outlines how the measurements of the two radioxenon monitors are performed. The various radioxenon sources in the Ottawa region are described in Section 3. Section 4 of this paper explains the data under study and the choices of events which were modelled. Meteorological modelling methods are illustrated in Section 5. Section 6 discusses the results of these methods and compares the results of the two different meteorological methods.

2. Measurements

2.1. NaI(Tl) network

The Ottawa Valley, NaI(Tl) network is composed of eight 7.62 cm diameter by 7.62 cm thick ($3'' \times 3''$) cylindrical NaI(Tl) crystals, facing skyward to measure radioactive plumes. Each detector is an Exploranium GR-150, and is more completely described in Grasty et al. (2001a,b). They are self-contained units consisting of a NaI(Tl) crystal, spectrometer, computer, data storage and modem and are built to withstand the harshness of Canadian winters.

The detectors take spectroscopic measurements every 15 min. The resulting measured spectrum is then divided into seven energy windows (argon, potassium, uranium, ^{133}Xe, ^{135}Xe, and others). For each energy window, a spectral stripping process is used to determine the number of counts per second in the particular window. A calibration coefficient is applied to convert the counts per second into a dose (Grasty et al., 2001b). The window which

Table 1
The half-lives of the four isotopes of radioxenon that the SPALAX measures (Nudat, 2006)

Isotope	Half-life
^{131m}Xe	11.934 days
^{133m}Xe	2.19 days
^{133}Xe	5.243 days
^{135}Xe	9.14 h

corresponds to ^{135}Xe overlaps with the major photopeak of ^{133m}Xe, so measurements in this window can belong to either of the isotopes ^{135}Xe or ^{133m}Xe.

The detectors recorded air kerma rate measurements in 15 min intervals and reported daily, but are able to alarm on a large signal in the case of an emergency. They are designed to measure the air kerma rate of ^{133,135}Xe and ^{41}Ar, but have also measured ^{99m}Tc. The detector has sensitivity to ^{133,135}Xe and ^{41}Ar at the level of less than 1% of ambient background.

They are calibrated monthly with the historical data of the detector in order to distinguish natural radioactivity from anthropogenic radioactivity. The detectors are gain stabilized by use of the ^{208}Tl gamma ray at 2614.7 keV (Grasty et al., 2001a).

The NaI(Tl) detectors give results as air kerma rate. For meteorological modelling, a conversion factor between air kerma rate and activity concentration must be used. This conversion factor between air kerma rate and activity concentration for ^{133}Xe was empirically found not to be the same as in ICRU 53 (1994). That conversion factor has been thoroughly investigated through Monte Carlo techniques (Stocki et al., 2007).

The detection limits (Table 2) for these NaI(Tl) detectors were taken from Grasty et al. (2001b) and then converted to daily detection limits using Eq. (6) of Grasty, where the 800 non-background samples per month were converted to 29 background samples per day. Then, by the use of air kerma to activity concentration conversion factors from ICRU 53 for ^{135}Xe and ^{41}Ar, these limits were converted into activity concentration limits. For ^{133}Xe the empirical number of 2.6 ± 0.2 pGy h^{-1} per Bq m^{-3} from Stocki et al. (2007) was employed.

2.2. SPALAX

Unlike the NaI(Tl) detectors, the SPALAX utilizes a High Purity Germanium (HPGe) detector. It samples the air daily and reports an activity concentration. The SPALAX is more completely described elsewhere (Fontaine et al., 2004; International Patent 1999).

The CTBT has a stringent set of requirements (CTBT prepcom document 1997; Schulze et al., 2000) for radioxenon monitoring. Four isotopes/isomers of radioxenon must be measured, namely ^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe. In particular, the ^{133}Xe minimum detectable concentration has to be less than 1 mBq m^{-3}. The half-lives of these isotopes/isomers are less than 12 days (see Table 1in Schulze et al., 2000), so the collection time for radioxenon has to be less than 24 h. The reporting frequency must be daily, which requires that the xenon collection frequency should the same. This high frequency of reporting implies that automation is necessary. The SPALAX satisfies all of these requirements.

The SPALAX extracts the four above mentioned radioxenon isotopes/isomers automatically and measures the activity concentration of each one. This air sampling is achieved by the use of a gas permeation membrane with a noble gas specific adsorbent used in conjunction. Sampling can be run continuously without the need for cryogenic cooling, external purge (to remove the xenon for the ovens), or a carrier gas supply. The SPALAX can extract and concentrate stable xenon by a factor greater than 10^6.

The SPALAX is designed to: maximize the efficiency of sampling and concentrating atmospheric radioxenon; minimize the radon concentration in the process; function continuously; function automatically; be reliable; detect ^{131m,133m,133,135}Xe simultaneously. All of this is done through four stages, namely (Fontaine et al., 2004):

(1) sampling,
(2) concentration and purification,
(3) ultimate concentration, and finally,
(4) detection.

The SPALAX was run with a 24-h sampling period followed by a 23.6 h period for spectrum acquisition, for the results reported in this paper.

Table 2 lists the detection limits from the SPALAX, calculated using the Aatami software (CTBTO Preparatory Commission, 2003), for a spectrum measured in Ottawa. These detection limits are measured in mBq, they were converted to mBq m^{-3}, via dividing by the SPALAX sample volume of 32 m^3. The 1 mBq m^{-3} ^{133}Xe minimum detectable concentration CTBTO requirement is fulfilled.[1]

3. The Ottawa environment

The Ottawa radioxenon environment is a complex, industrialized setting due to the significant amount of nuclear power production and nuclear industry (Stocki et al., 2005). Ottawa is unique in its proximity to the largest producer of medical isotopes via uranium fission in the world. Potential sources of emissions of radioactive noble gasses progressing from the local, to the more regional scale include:

(1) hospitals that use radioisotopes in Ottawa;
(2) a commercial and research facility operated by MDS Nordion in the west end of Ottawa (MDS Nordion web site);
(3) a research reactor operated by Atomic Energy of Canada Ltd. in Chalk River, approximately 180 km to the north-west of Ottawa;
(4) a medical isotope manufacturing facility at the same Chalk River site operated by AECL–CRL (Atomic Energy of Canada Ltd and Chalk River Laboratories) for MDS Nordion and capable of producing more than 100% of the world's requirements of ^{133}Xe via the ^{235}U fission;
(5) a number of CANDU nuclear power generating (NPG) stations to the south-west, north-east and east of Ottawa from 300 to 700 km in distance;
(6) at some distance beyond the Canadian NPG stations, there are several nuclear power reactors in the United States of America;
(7) ^{131}I is also produced at the Chalk River site via neutron capture on ^{130}Te. This is a potential source of ^{131m}Xe from ^{131}I decay.

The largest source of these radioisotopes is number 4 above with a normal daily ^{133}Xe emitted activity which, as an example, can be ranging between 5 and 44 TBq/day (Saey et al., 2007). The second largest source is the nuclear power reactors which are a few orders of magnitude smaller (Kalinowski et al., 2005). The distributions of these emissions will be subject of another paper.

Fig. 1 shows the wind direction frequencies (on a short mast on top of building 456 at Chalk River Laboratories) indicated by the wind rose, produced by data from Niemi et al. (2001). One can see that the wind blows in the direction from Chalk River to Ottawa 15% of the time. The area encompassing Chalk River Laboratories is well equipped. On the site, a meteorological mast is capable of measuring local temperature, wind speed and wind direction.

4. Available measurements

4.1. NaI(Tl) network measurements

The first set of detectors in the Health Canada network has been providing measurements since the summer of 2002. In this paper, we have selected certain days in which there was a significant

[1] A new version of the SPALAX with improved performance has been developed and industrialized by Environment SA and is now available.

Table 2
Detection limits for the NaI(Tl) detectors and the SPALAX

Isotope	NaI(Tl) detector				SPALAX	
	Single spectrum detection limit (nGy per 15 min)	Daily detection limit (nGy day^{-1})	Single spectrum detection limit (mBq m^{-3})	Daily detection limit (mBq m^{-3})	Daily minimum detectable activity (mBq)	Daily minimum detectable concentration (mBq m^{-3})
^{133}Xe	0.01	0.05	15,000	800	20	0.63
^{135}Xe	0.025	0.13	1800	100	62	1.94
^{41}Ar	0.1	0.54	1300	74	n/a	n/a
Skyshine	0.075	0.40	n/a	n/a	n/a	n/a

The first column is from Grasty et al. (2001b).

plume traveling down the Ottawa Valley. The data were analyzed with Exploranium software.

In some unusual cases, the amount of xenon traveling down the valley has been large enough to be detected by the NaI(Tl) network. These were easily identified and meteorologically modelled for the dates shown in Fig. 2.

Fig. 2 indicates that the June 5, 2003 event might have been due to medical grade xenon, i.e., it was a product about to be shipped.

The NaI(Tl) network data have shown that xenon plumes, apparently originating from Chalk River, move down the valley to Ottawa, once or twice per month. There tends to be a correlation between these plumes and the production of ^{99}Mo.

To better understand the source of these radioxenon plumes, one must have some background about the extraction of ^{99}Mo. In the ^{99}Mo recovery process (IAEA, 1998) the target is allowed to cool, and the irradiated target cell is stripped in a hot cell to remove the external cladding. Then it is dissolved in nitric acid, producing ^{133}Xe in the off-gas stream. Radioactive noble gases are also emitted during the cementing process of high-level liquid wastes (Thomson, 2004). Both the dissolution and cementing processes result in short term intermittent releases. These releases can be considered "medical isotope production" releases of xenon. In 2003, there were no radiation exposures in excess of Action Levels (Thomson, 2004, see p. 1). These releases constitute a small fraction of the Derived Release Limit, and are below Action Levels (Thomson, 2004, see p. 1) and are in compliance with Canadian Nuclear Safety Commission regulations.

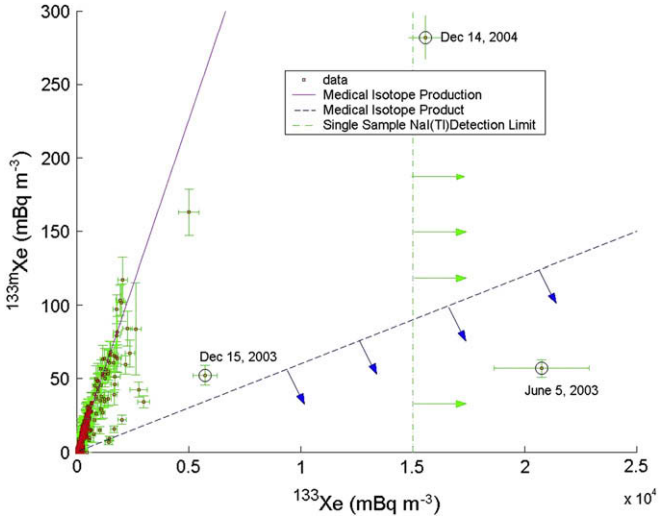

Fig. 2. Activity concentration measured by the SPALAX in Ottawa for ^{133m}Xe vs. ^{133}Xe. Indicating which data point, where the temporal information from the NaI(Tl) network was considered for meteorological modelling. It appears that two of the dates are close to the medical isotope product line, implying they might be medical isotope product. The "medical isotope product" line on was generated from the MDS Nordion specification (see the Nordion web site reference). The other one was most likely waiting to be aged to become product. Note that none of these days are from the medical isotope production data set as it is far from the single spectrum NaI(Tl) detection limit.

To give a sense of scale for the source terms which will be discussed, up to 1 GBq of ^{133}Xe can be administered internally to patients in radiodiagnostic procedures (DraxImage web site). This results in a dose on the order of 0.27–0.45 mGy. The composition of xenon observed in Ottawa was completely consistent with medical grade radioxenon. Hence, the dose to any individual resulting from these plumes likewise scales to the medical procedure in proportion to the amount of xenon used in radiodiagnostic procedures.

4.2. SPALAX measurements

The data presented here are from measurements taken from the roof top at the Radiation Protection Bureau Building in Ottawa, from June 20, 2001 to May 11, 2004. The sampling period was 24 h and 632 measurements were made. This station detected ^{133}Xe almost every day that the SPALAX was running. The data were analyzed using the Genie 2k software developed by Canberra Industries.

Fig. 2 is a graph of activity concentration from the SPALAX. There are three data points indicated on the graph with their dates. Some of the interesting SPALAX data are on a line which corresponds to a ratio of ^{133m}Xe to ^{133}Xe of 0.045. This ratio corresponds to medical isotope production (Stocki et al., 2005). Fig. 2 and Table 2 show that this line lies below the single spectrum detection limit of ^{133}Xe for the NaI(Tl) detectors: the maximum value of ^{133}Xe on that line is 2.64 ± 0.24 Bq m^{-3} whereas the single spectrum detection limit for ^{133}Xe for the NaI(Tl) detector is 15 Bq m^{-3}.

4.3. Choice of events under study

According to Fig. 2 there are three events which are very interesting measurements made by the SPALAX in terms of quantity of ^{133}Xe and in terms of their ^{133}Xe/^{133m}Xe isotope ratio. These three points are excellent candidates for meteorological modelling. The June 5, 2003 event was pioneering work, which began the investigations in this publication. This paper will focus on the releases which correspond to December 15, 2003 and June 5, 2003. The date December 14, 2004 is equally interesting.

Another interesting study involved finding an event that had been seen by the NaI(Tl) network detectors and had the medical isotope ratio ^{133m}Xe to ^{133}Xe of 0.045. A detailed study of the top 10 events on that medical isotope line was performed to find the best candidate. The event had to be seen by a large number of detectors in the NaI(Tl) network (measuring both ^{133}Xe and ^{135}Xe); have a recognizable signal at the Ottawa location; and have a high signal-to-noise ratio in both ^{133}Xe and ^{135}Xe measurements. The potential of artifacts due to the uranium in the signal was also investigated. An attempt to model one of these types of events was unsuccessful; which is not surprising as the NaI(Tl) signals were very weak and below the theoretical detection limit shown in Table 2. These measurements of radioxenon by the SPALAX which correspond to a ratio of ^{133m}Xe to ^{133}Xe of 0.045 could also be quasi-continuous or continuous releases, unlike the discrete releases described in this paper.

5. Meteorological modelling description

Xenon is a noble gas. For atmospheric transport and dispersion it behaves like a simple inert tracer, unlike other tracers which have complex, physical and chemical behavior and may be subject to wet scavenging and dry deposition.

Two radioxenon events which were observed by the NaI(Tl) network and the SPALAX, were modelled by two separate institutions: the Canadian Meteorological Centre (CMC) and the Commissariat à l'Energie Atomique (CEA) – Département Analyse, Surveillance, Environnement (DASE). The descriptions of the models are presented below. For a more general explanation of various models of dispersion of radionuclides in the atmosphere (see for e.g. Cooper et al., 2003).

In the time periods of June 4–7, 2003 and December 11–19, 2003, the four xenon radioisotopes, ^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe, were detected by the SPALAX installed at Health Canada – RPB in Ottawa and operated with a 24 h-sampling time. During both periods, the average ^{133}Xe activity concentration was less than 1 Bq m^{-3}, except on

- 12:00 h UTC on June 5, 2003 to 12:00 h UTC on June 6, 2003 with a ^{133}Xe detection event of more than 20 Bq m^{-3},
- 12:00 h UTC on December 15, 2003 to 12:00 h UTC on December 16, 2003 with a ^{133}Xe detection event of more than 5 Bq m^{-3}.
- 12:00 h UTC on December 14, 2004 to 12:00 h UTC on December 15, 2004 with a ^{133}Xe detection event of more than 15 Bq m^{-3}.

These three events are summarized in Table 3.

Due to their relative closeness (approximately 180 km up the Ottawa Valley from RPB), Chalk River Laboratories were identified as the most likely source.

5.1. The Canadian Meteorological Centre (CMC) models

In this study two dispersion models were used to model the NaI(Tl) data: a short range dispersion model and a long range dispersion model. This is unlike Stocki et al. (2005) where a simple back trajectory model was used. The short range model, Modèle Lagrangien à Courte Distance (MLCD), was used to obtain a rough estimate of the source term, from nearby dose rate measurements and local meteorological observations. The source location was assumed to be the production facility at the Chalk River Labs (CRL). MLCD has been developed for local scale emergency response (for within 10 km of the spill site, but in certain conditions it can be applied to ranges up to 50 km from the source of emission). It is a three dimensional Lagrangian particle model which is based on a Langevin stochastic equation for turbulent diffusion. The model can be run in forward or inverse mode. The model assumes horizontal uniformity of the meteorological conditions. A more thorough description of MLCD can be found in Flesch et al. (2002).

The source term was then used with a long range model (Modèle Lagrangien de Dispersion de Particules d'ordre Zéro (MLDP0; D'Amours et al., 2004)) to estimate the plume transport and dispersion at large distance. MLDP0 is also a three dimensional Lagrangian particle model. For vertical mixing the model is based on a random displacement equation with a diffusion coefficient. MLDP0 is an off-line model and meteorological conditions are provided by the Global Environmental Multiscale GEM (Côté et al., 1998a,b) modelling system running operationally at the CMC. MLDP0 can also be run in forward and inverse modes.

5.2. The Commissariat à l'Energie Atomique (CEA) models

5.2.1. Meteorological fields

The meteorological fields were simulated with the MM5 (Mesoscale Model 5th generation; Dudhia, 1993; Grell et al., 1994; Dudhia et al., 2003) and WRF (Weather Research and Forecasting) suites developed at Pennsylvania State University and National Center for Atmospheric Research. MM5/WRF are limited area modelling systems used to solve the non-hydrostatic compressible equations of the atmosphere dynamics on increasing resolution nested domains. The calculations were performed in 'two-way nesting' mode.

5.2.1.1. Regional study. Low resolution simulations were carried out with MM5 and WRF on two to three nested grids. The mesoscale and regional grids covered respectively an area of 700 km × 700 km with a resolution of 13.5 km × 13.5 km and an area of 350 km × 350 km, with a resolution of 4.5 km × 4.5 km. This second grid was centered on Chalk River. A third grid was used in the final simulations.

5.2.1.2. Local study. High resolution simulations were performed with MM5 on five nested grids with horizontal resolutions ranging from 81 km to 1 km. The coarsest resolution grid covered the north-east portion of America. The finest resolution grid zoomed in on Chalk River Laboratories and vicinity. The five-domain vertical grid had 27 levels between the soil and the 100 hPa pressure level. The time step of the grid with the coarsest resolution was 240 s.

The quality of the simulated wind and temperature was assessed by comparing MM5 results with the observations at Chalk River Laboratories meteorological mast. The agreement of the calculations with the measurements was excellent, demonstrating that the high MM5 space and time resolution was capable of reproducing the rapid variations in the speed and direction of the wind. These accurate meteorological fields warrant the precision of the dispersion calculations.

5.2.2. Dispersion

The atmospheric dispersion was simulated with FLEXPART, a Lagrangian code able to deal with nested domains, developed at Munich University by Stohl et al. (1998). FLEXPART was integrated forward and backward in time to compute the trajectories of numerous particles representing radioxenon parcels released by

Table 3
The levels and ratios of Xe measured by the SPALAX in Ottawa for the observations, in which the temporal data from the NaI(Tl) network were considered for meteorological modelling

	June 5, 2003 observation			December 15, 2003 observation			December 14, 2004 observation		
	Bq m^{-3}	Uncertainty (Bq m^{-3})	% ^{133}Xe	Bq m^{-3}	Uncertainty (Bq m^{-3})	% ^{133}Xe	Bq m^{-3}	Uncertainty (Bq m^{-3})	% ^{133}Xe
^{133}Xe	20.8	2.1		5.74	0.53		15.6	0.8	
^{133m}Xe	0.0569	0.0058	0.27	0.0522	0.0067	0.90	0.282	0.015	1.80
^{135}Xe	0.006	0.001	0.03	0.228	0.022	3.87	0.344	0.096	2.21
^{131m}Xe	0.168	0.021	0.81	0.0421	0.0074	0.73	0.063	0.005	0.40

The uncertainties are at the 1σ level.

the source and transported by the mean and turbulent wind components.

For both the regional and local studies, MM5/WRF meteorological fields stored in every hour were used as input data in FLEXPART. The particles were emitted from a point source, centered at 50 m above the ground level. The activity concentration field was computed on a 3D grid similar to the finest MM5 grid. It was averaged on a time period of 900 s in the local study, and of 3600 s in the regional study.

In the regional study, numerical tests using 10,000–500,000 particles showed that 50,000 particles were sufficient to propagate a plume over a distance of around 180 km (distance between Ottawa and Chalk River Laboratories). The local study took account of ^{133}Xe radioactive decay ($\tau_{1/2} = 5.25$ days), while there was no significant dry, or wet, deposition of ^{133}Xe.

6. Meteorological modelling data analysis

6.1. The December 15, 2003 xenon event (CMC results)

On December 16, 2003 a 24-h average ^{133}Xe concentration in excess of 5 Bq m^3 was detected by the Ottawa SPALAX. This measurement was clearly related to a significantly elevated background dose rate detected by the co-located NaI(T1) monitor (Fig. 3 top graph), between 21:00 h UTC, on the 15th, and 00:00 h UTC on the 16th. The event also appears related to dose rates measured by the detectors at Sheenboro, Chapeau, and Westmeath a few hours earlier. Surface winds reported at the Petawawa and Ottawa airports during the period were generally from the north-west around 5 m s^{-1}; this supports the hypothesis of a simple transport of a xenon plume originating from CRL along the axis of the stations (Fig. 1). Inverse calculations were done with the MLCD using the measurements in Sheenboro, together with the Perch Lake Tower wind observations. Based on the modelling results and on the interpretation of qualitative in-stack radiation monitoring, a source term was approximated by a continuous release on the order of 10^{14} Bq over 6 h, starting at 13:00 h UTC on the 15th. This source term was provided to the long range model MLDP0, which was executed using meteorological data supplied by the regional GEM data assimilation system in operation at the CMC in 2003. The results of this forward simulation are shown in Fig. 3 (bottom). The model reproduces relatively well the temporal behavior of the plume in Sheenboro; the approximate shape of the plume in Chapeau, and the double peak shape of the plume in Westmeath are also reproduced. The modelled plume does not reach the RPB samplers (Fig. 4); however concentrations calculated at a virtual sampler just 10 km to the west of RPB do reproduce fairly well those observed in Ottawa. This gives a measure of the uncertainties associated with the modelling. Indeed, even though the atmospheric circulation was fairly constant from the north-west and apparently not strongly influenced by the topography, slight shifts in wind direction can mean that a relatively thin plume will be missed.

6.2. The June 5, 2003 xenon event (CMC results)

On June 5, 2003, the SPALAX system in Ottawa observed ^{133}Xe in excess of 20 Bq m^{-3}. This event was the largest observed ^{133}Xe measured in the Ottawa area, and is the pioneering work that started the investigation into meteorological modelling of radioxenon events down the Ottawa Valley. Table 3 lists the levels and ratios of the isotopes observed by the SPALAX in Ottawa. Fig. 5 shows the amount of air kerma (measured in nGy h^{-1}) due to ^{133}Xe measured at Petawawa, then in Chapeau, and then at Ottawa from the release on June 5, 2003. Clearly a significant xenon plume was moving south-east from Chalk River in the direction of Ottawa. However the dose rate measured at Sheenboro, which lies directly on the axis CRL-Chapeau-RPB is much less than what is observed in Petawawa or Chapeau, and the timing is almost synchronous with Chapeau while about halfway between the two. This indicates a more complex transport than a simple smooth translation of the plume down the valley to Ottawa.

The nearest NaI(Tl) detector observation (and also the nearest population centre) to the medical isotope facility was taken some 30 km from the site. The peak dose rate, lasting no more than an hour, was 6 nGy h^{-1}. This represents about a 25% increase in external dose rate compared to average background but is well within normal variation of background at this location.

The dose rates observed in Petawawa were converted to air concentrations using an empirical calibration resulting from the comparison of the Ottawa SPALAX observation and those of the co-located NaI(Tl), for the case. MLCD was run in inverse mode using wind observations from the Perch Lake (which is 1.4 km away from Chalk River Laboratories) meteorological tower and from the weather station in Petawawa to estimate, using the derived concentrations, a source strength of about 5×10^3 TBq, assuming that the source was located at the Chalk River production facility.

That source strength, with an assumed emission period from 16:00 h UTC to 18:00 h UTC on June 5 was then used as an input for MLDP0 to simulate the plume dispersion down the Ottawa Valley. Meteorological conditions for the dispersion model were taken from the CMC operational GEM regional data assimilation system available at the time at a horizontal resolution of 24 km. Fig. 6 shows the coherence between the SPALAX and NaI(Tl) measurements that is reported in other studies (Stocki et al., 2007).

The agreement between the modelled and observed concentrations is quite close, especially with regard to the timing of the plume passage, and supports the hypothesis of a fairly large release. However, given uncertainties associated with the wind fields and the importance of local effects for short range dispersion – also given that the long range model is unable to satisfactorily reproduce the observations at close range, the estimated source term could be in error by one or two orders of magnitudes.

6.3. The December 15, 2003 xenon event (CEA results)

6.3.1. Regional study (SPALAX detection)

A regional numerical study was undertaken to explain the 5.74 Bq m^{-3} detection of ^{133}Xe by the Ottawa SPALAX on December 15, 2003. Low resolution meteorological fields were simulated with MM5 on two nested grids (see Section 5.2.1). In the mesoscale grid, the calculated horizontal winds, temperature and relative humidity were nudged towards 6-hourly time wind fields from NCEP reanalysis.

Time and wind were inverted in order to compute the ^{133}Xe backward transport with FLEXPART and determine the origin of the air parcels with ^{133}Xe arriving in Ottawa (see Section 5.2.2). ^{133}Xe was considered as an inert tracer and a unit activity was released from the SPALAX location on 12:00 h on December 16, 2003 for a 24 h-period back in time. The backward activity concentrations at the grid points represent dilution coefficients from the receptor to potential sources. These coefficients are inversely proportional to the intensity of the potential source.

From the numerical simulations, the ^{133}Xe retroplume (plumes calculated backward in time by dispersion models) stagnated above Ottawa during the first 5 h. It propagated (backward in time) then in the north-west direction from 06:00 h on December 16 to 12:00 h on December 15, 2003 and affected the Chalk River vicinity between 20:00 h and 18:00 h on December 15, 2003. This is consistent with the real situation of the plume starting in Chalk River and then heading south-east to Ottawa and lingering in Ottawa. Fig. 7 shows the backward activity

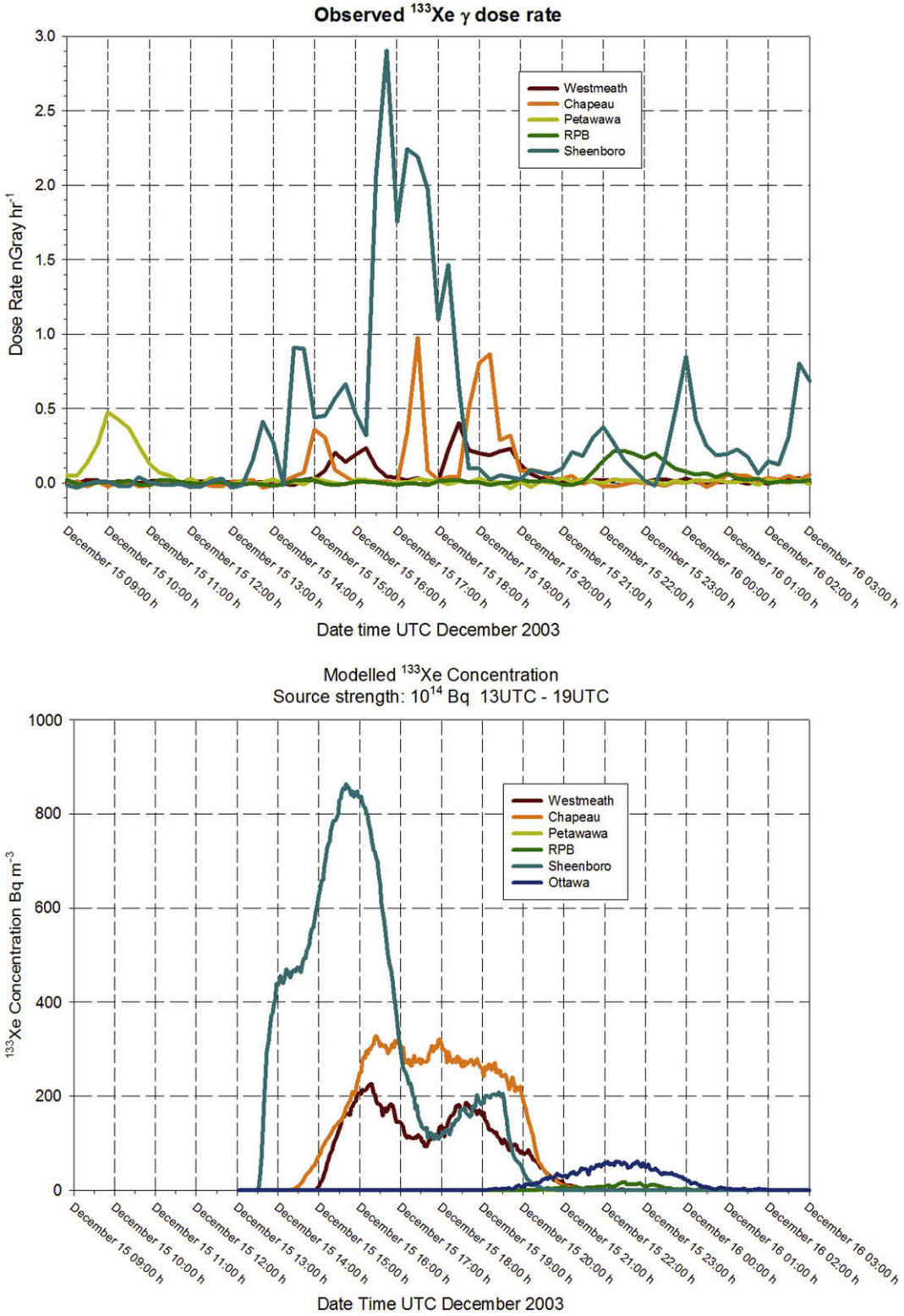

Fig. 3. Top: December 15, 2003 measurements of ^{133}Xe from the Ottawa Valley sub-network. Bottom: The environmental model of the measurements. Stations indicated in different colours: Westmeath (red), Chapeau (orange), Petawawa (gold), Radiation Protection Bureau, Ottawa, (green), Sheenboro (light blue), virtual Ottawa (blue).

concentration field averaged over 1 h on December 15, 2003 at 24:00 h, 22:00 h, 20:00 h and 18:00 h (UTC). Chalk River is situated at the edge of the retroplume. The 1-h-average dilution coefficient in Chalk River Laboratories was about 1×10^{-13} m^{-3} at 19:00 h on December 15, 2003. Thus, a release of 5×10^{13} Bq on this date and over a time period of 1 h from Chalk River Laboratories accounts for the recorded peak of about 5 Bq m^{-3} in Ottawa.

Complementary transport simulations in direct mode from Chalk River Laboratories to Ottawa were carried out with a source

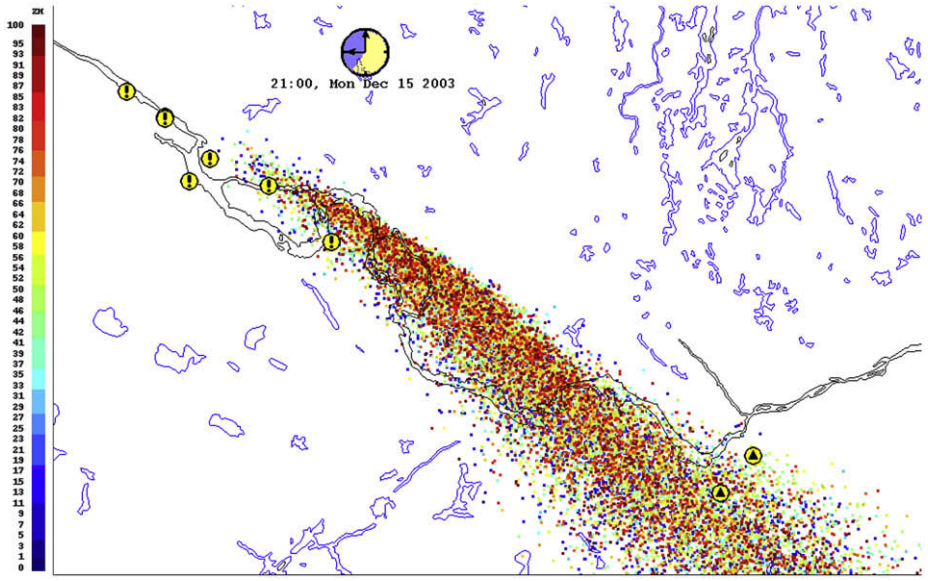

Fig. 4. Position of modelled air parcels of the December 15, 2003 event, at 21 UTC. The icons with ! marks indicate the position of NaI(T1) detectors in the vicinity of the Chalk River Labs, those with triangles location of the Ottawa RPB NaI(Tl) detector (East) and the virtual NaI(Tl) detector (West) where concentrations were also calculated (see text). The location of this "virtual detector" is an indication of horizontal uncertainty.

emitting between 18:00 h and 19:00 h on December 15, 2003. They show that the ^{133}Xe plume propagates south of Ottawa and confirms the order of magnitude of the potential source which is also compatible with the NaI(Tl) signals recorded at Sheenboro, Chapeau and Westmeath between 18:00 h and 19:00 h on December 15, 2003. The absence of NaI(Tl) signal in Petawawa as well as the signal recorded later at RPB could be due to their southern locations.

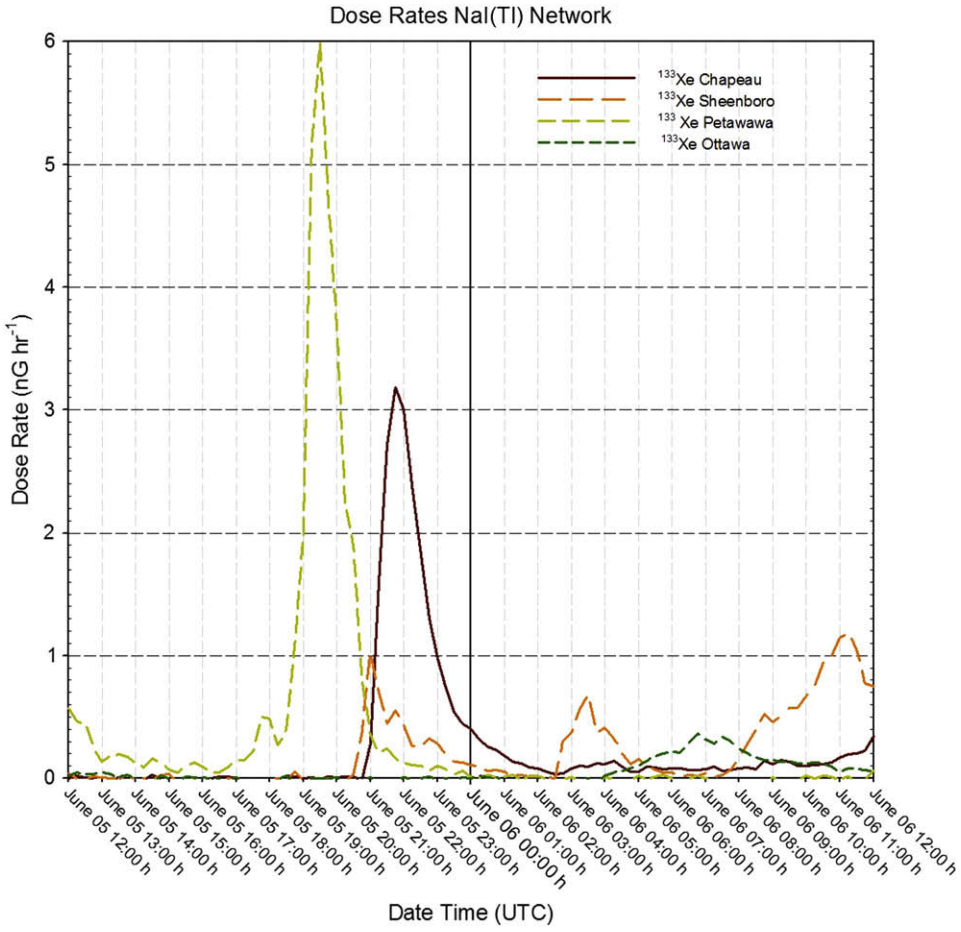

Fig. 5. Release on June 5, 2003. A ^{133}Xe plume traveling down the valley and measured at 3 of the NaI(Tl) network detectors. First in Petawawa (yellow dash), then in Chapeau (continuous red), and finally in Ottawa (long dashed green).

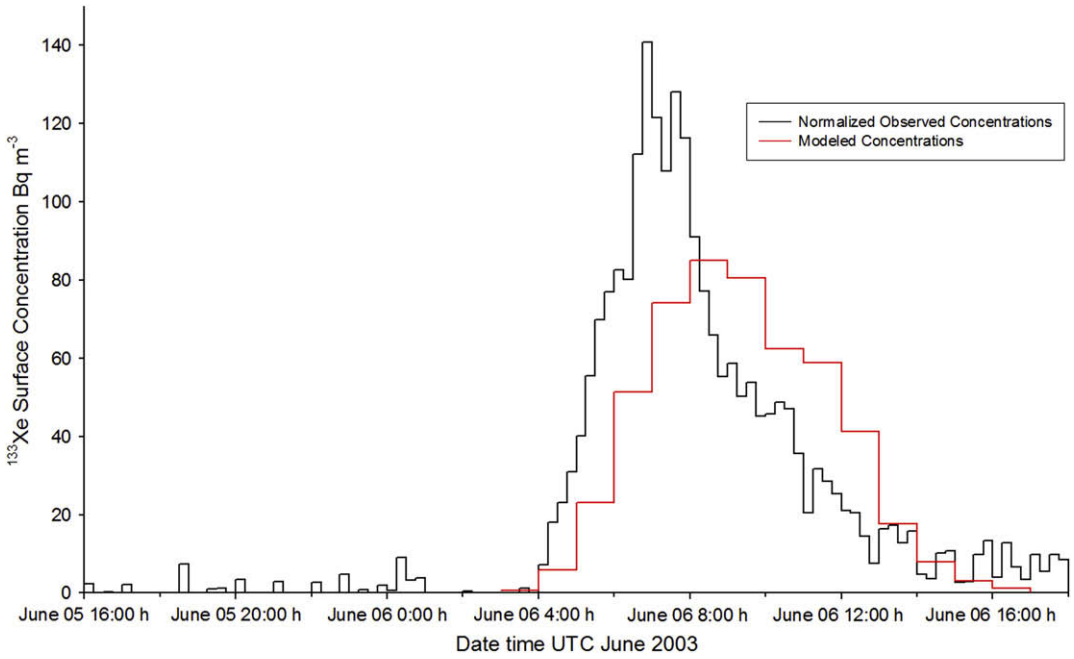

Fig. 6. In black, data measured by the NaI(Tl) in Ottawa normalized to the SPALAX collocated in Ottawa. In red, MLDP0 modelled concentration assuming a release of 5×10^{15} Bq in Chalk River from 16 UTC to 18 UTC June 5, 2003.

Our results are confirmed by downwind dilution factors found in the UNSCEAR (Annex A, Table V, 2000) report. These coefficients are based on Gaussian plume model calculations. For a distance of 200 km, they are about 3×10^{-10} s m^{-3} or 10^{-13} h m^{-3}, which is the dilution factor calculated in this regional study.

6.3.2. Local study (NaI(Tl) detections)

A local numerical study was also performed taking into account all the measurements on the NaI(Tl) detectors in the vicinity of Chalk River. As the most probable xenon source was Chalk River Laboratories, the study did not aim at diagnosing the location of the

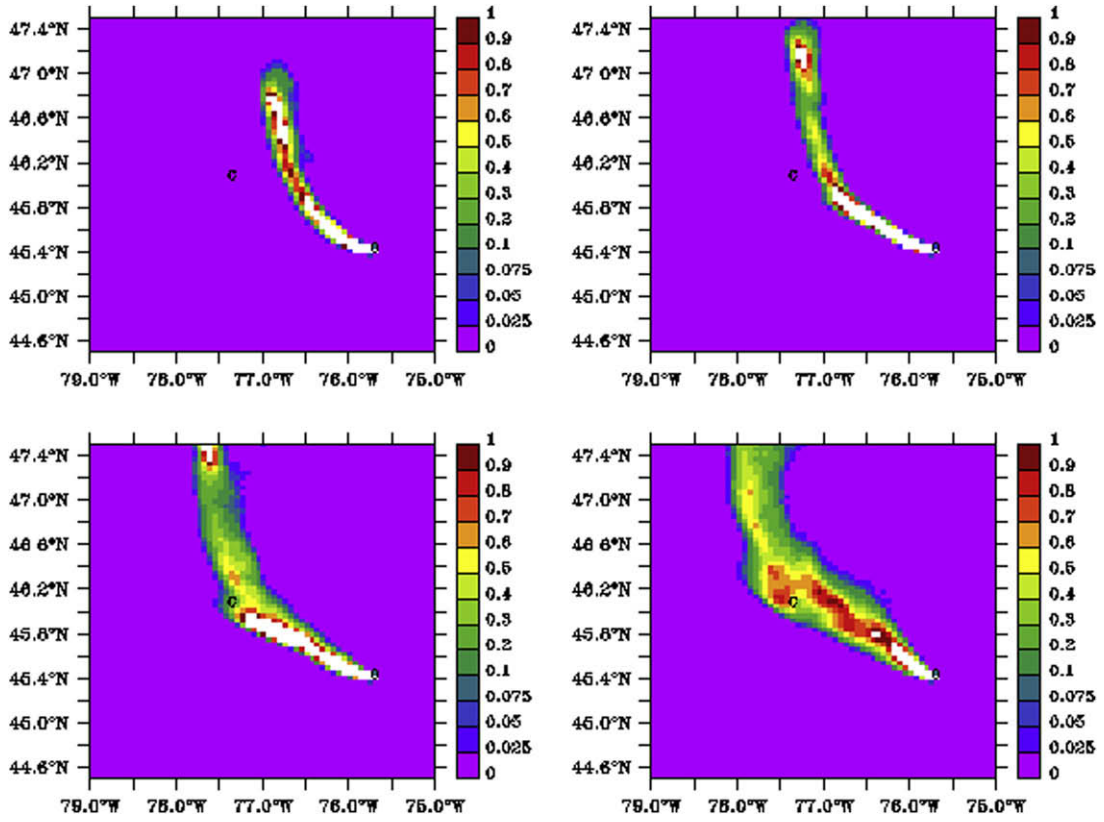

Fig. 7. Backward activity concentration (or dilution coefficient) field for a unit release by the SPALAX in Ottawa, from 12:00 h on December 16, 2003 to 12:00 h on December 15, 2003 (UTC). The dilution coefficient is averaged over 1 h and scaled by 1×10^{13}. Chalk River Laboratories and Ottawa are denoted by the letters C and O, respectively.

source but focused on the kinetics and magnitude of the ^{133}Xe emissions.

The time histories of ^{133}Xe activity concentration measured by the NaI(Tl) detectors are presented in Fig. 8 for the period of 00:00 h on December 15, 2003 to 00:00 h on December 17, 2003. The maximum ^{133}Xe activity recorded by Deep River and Sheenboro (the nearest detectors to Chalk River Laboratories) is more than 1000 Bq m^{-3}. As the detectors are located respectively north-west and south-east of Chalk River, it indicates the wind reversed on December 16, 2003 around 09:00 h; contrary to what the Deep River and Sheenboro, Chapeau, and Petawawa detectors (only 20 km south-west of Sheenboro) measured which was a weak ^{133}Xe activity. This illustrates the variability of dispersion at the local scale and the necessary high resolution wind predictions to take account the local effects. The time histories of ^{133}Xe activity concentration also demonstrate that a short release cannot be responsible for all the NaI(Tl) detections. In fact, a more or less continuous emission evolving with time is more plausible for the studied period.

High resolution simulations of the meteorological flow and ^{133}Xe transport were carried out with MM5 and FLEXPART modelling system (see Sections 5.2.1 and 5.2.2). An 'inversion method' was applied to the measurements issued by the NaI(Tl) detector network. The principle is as follows. Eighteen successive runs of FLEXPART were performed considering continuous releases with a 3-h duration and a magnitude of 1 Bq. For each FLEXPART run, the calculated ^{133}Xe activities on six receptors corresponding to the NaI(Tl) detectors were stored with a time step of 15 min. The 'real' temporal evolution of the source term was computed taking account of the measurements on the NaI(Tl) detectors. As this kind of problem is mathematically 'ill-posed' within the meaning of Hadamard (the solution for the source term exists, but is not unique and not continuous), a 'regularization method' was introduced in order to reveal the "best" continuous solution. In this study, Tikhonov and Backus–Gilbert regularization methods were used (Hansen, 2005; Press et al., 1992) and led to comparable results.

Fig. 9 shows the calculated estimation of the amplitude of the radioxenon releases by Chalk River Laboratories. This was computed using MM5 wind fields (including a nudging towards ECMWF analyses and local meteorological data), FLEXPART dispersion code, and Tikhonov regularization method. Comparable results were obtained with slightly different MM5 wind fields (relaxation towards NCEP/NOAA (National Center for Environmental Prediction/National Oceanic and Atmospheric Administration) analyses, no observational nudging) and Backus–Gilbert regularization. Finally, the calculated radioxenon emissions were as follows:

- A first release of magnitude 1×10^{13}–8×10^{13} Bq took place between 15:00 h and 18:00 h on December 15, 2003. It was responsible for the detections at Chapeau, Sheenboro and Westmeath the same day between 16:00 h and 20:00 h, and for the detection in Ottawa between 22:00 h and 23:00 h.
- A second release of magnitude 3×10^{13}–6×10^{13} Bq was emitted between 00:00 h and 06:00 h on December 16, 2003. It was responsible for the high detection at Sheenboro the same day between 03:00 h and 09:00 h.
- A third release of magnitude 1×10^{13}–2×10^{13} Bq took place between 09:00 h and 12:00 h on December 16, 2003. It was responsible for the detection at Deep River the same day between 10:00 h and 12:00 h.
- A fourth release of magnitude 3×10^{13}–6×10^{13} Bq was emitted between 15:00 h and 18:00 h on December 16, 2003. It was responsible for the second detection at Deep River the same day between 16:00 h and 19:00 h.

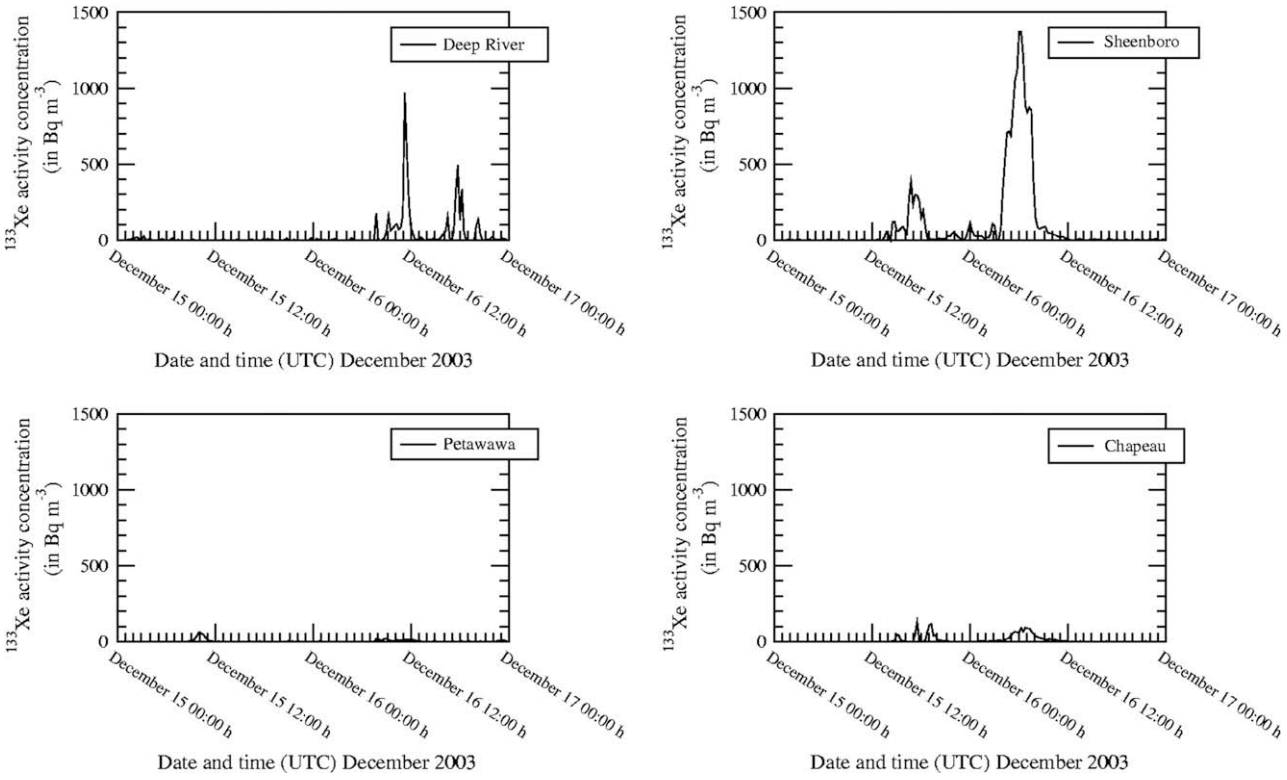

Fig. 8. Time histories of ^{133}Xe activity concentration measured by the NaI(Tl) detectors located down the valley of the Ottawa River. The time period is 00:00 h on December 15, 2003 to 00:00 h on December 17, 2003. There are seven NaI(Tl) detectors: Deep River, Sheenboro, Petawawa, Chapeau, Westmeath, and Ottawa – RPB, but only the time histories of four of them with the highest signals are represented.

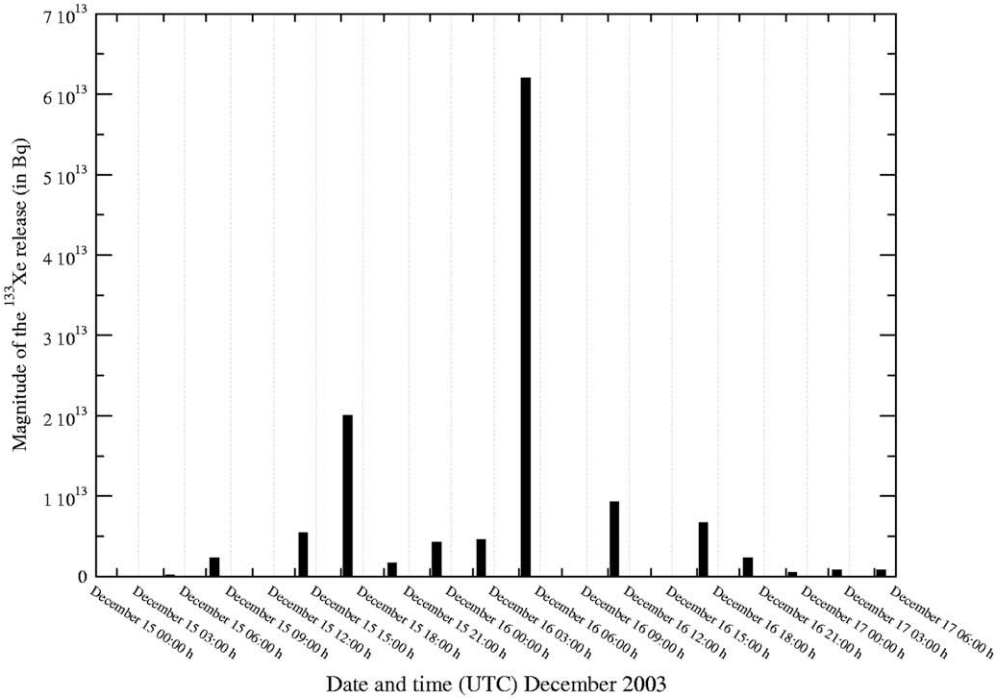

Fig. 9. Evolution with time of the magnitude (in Bq) of the releases by the Chalk River Laboratories. Simulations are based on MM5 wind fields (including a nudging towards ECMWF analyses and local meteorological data), FLEXPART dispersion code and Tikhonov regularization method.

The order of magnitude of the calculated releases (1×10^{13}–8×10^{13} Bq) is in accordance with realistic emissions in normal operation from Chalk River Laboratories. This validates the meteorological flow and dispersion modelling system, and also the methodology utilized to account for the NaI(Tl) detectors measurements.

6.4. The June 5, 2003 xenon event (CEA results)

6.4.1. Regional study (SPALAX detection)

Regional simulations were carried out to determine the origin of the ^{133}Xe peak of 20 Bq m^{-3} recorded by the Ottawa SPALAX from 12:00 h on June 5 to 12:00 h on June 6, 2003. From numerical simulations, the meteorological situation in the area was characterized by a low located south of Ottawa, moving eastwards and weakening. Winds from June 4–6 in the concerned area turned from east to north-west at the crossing of this low, as confirmed by meteorological measurements at Chalk River.

In order to better capture local phenomena, higher resolution simulations were performed using WRF model on three nested grids. The mesoscale and regional grids are those described in Section 5.2.1. The third grid uses a resolution of 900 m and covers an area of 200 km $\times$ 110 km including Chalk River and Ottawa (Fig. 10). Wind fields were guided by NCEP winds in the mesoscale grid only and were calculated in the three interacting grids over a period of four days starting on 00:00 h on June 3, 2003.

Retroplumes calculated from fine resolution simulations are averaged over 1 h and presented in Fig. 10 on June 6, 2003 at 06:00 h, 05:00 h, 04:00 h and 02:00 h (UTC). The retroplume immediately left Ottawa on June 6 at 12:00 h (UTC). It propagated in the west direction and then followed the river topography propagating in the north-west direction towards Chalk River. The retroplume reached Chalk River vicinity, 6 h after the retroemission at Ottawa and over a period of 4–5 h. The maximum concentration occurred around 9 h after the retroemission, i.e. on June 6 at 03:00 h. From 02:00 h, winds were turning east in this area and the retroplume from Ottawa propagated eastwards, without reaching Chalk River. Nevertheless, due to this wind direction reversal, a part of the retroplume located in the west of Chalk River went back in the east direction and propagated over Chalk River from 20:00 h to 16:00 h on June 5. During this latter time period, concentrations in the vicinity of Chalk River were about 10 times smaller that those calculated 9 h after the retroemission.

The retropropagation is consistent with direct propagation from Chalk River. Transport simulations in direct mode from Chalk River to Ottawa were carried out with a source emitting between 02:00 h and 03:00 h on June 6, 2003. They have shown that the ^{133}Xe plume propagated in the south-east direction, reaching Petawawa and Chapeau, 2 and 3 h respectively after the emission, and affecting Ottawa from 08:00 h to 12:00 h (UTC) on June 6. Taking into account NaI(Tl) measurements at Petawawa, Chapeau, and Ottawa, the signals of the calculated concentrations were shifted by 6–8 h. This time shift could be associated with the extracted global NCEP winds, which have guided our calculations in the mesocale grid and which turned from the east to the north-west on June 6 at 00:00 h (UTC). This change of wind direction occurred 6–8 h before, when analyzing ECMWF winds and around twelve hours before when considering wind measurements at Chalk River, compared to NCEP analyses. Considering this time shift, the emission would have occurred on June 5 in the afternoon. In this case, the plume would have propagated first westwards, turned back in the afternoon and then propagated towards Ottawa.

In view of the first hypothesis (calculations guided by NCEP winds), the calculated dilution coefficient between Ottawa and Chalk River was around 2×10^{-13} m^{-3} at 02:00 h (UTC) on June 6. So, the source intensity at this date and time, over 1 h had to be around 10^{14} Bq to obtain 20 Bq m^{-3} at Ottawa. Using ECMWF winds, a potential source emitting between 18:00 h and 20:00 h on June 5, would be obtained. Considering this second hypothesis, the intensity of the source should be 10 times higher.

Due to this complex meteorological situation, the regional study cannot discriminate between these two hypotheses. This event requires finer analysis, taking into account NaI(Tl) measurements.

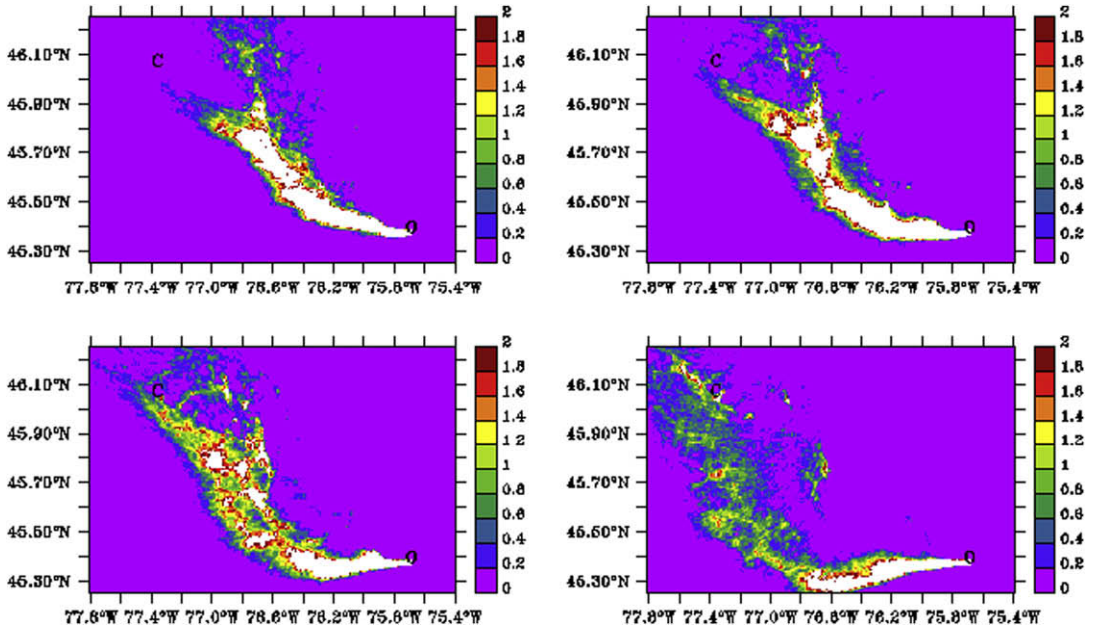

Fig. 10. Backward activity concentration field for a unit release by the SPALAX in Ottawa, from 12:00 h on June 6, 2003 to 12:00 h on June 5, 2003 (UTC). The dilution coefficient is averaged over 1 h and scaled by 1×10^{13}. Retroplumes are presented at 06:00 h, 05:00 h, 04:00 h and 02:00 h on June 6.

6.4.2. Local study (NaI(Tl) detections)

The local numerical study takes into account the measurements on the NaI(Tl) detectors in the vicinity of Chalk River as a whole. The xenon source is assumed to be located at the Chalk River Laboratories. The local simulations are carried out to determine the kinetics and magnitude of the ^{133}Xe emissions.

The times histories of ^{133}Xe activity concentration measured by the NaI(T1) detectors are presented in Fig. 11 for the period of 00:00 h on June 4, 2003 to 00:00 h on June 8, 2003. The ^{133}Xe activity recorded by Deep River is around 100 Bq m^{-3} and more than 700 Bq m^{-3} in Petawawa. As the detectors are located respectively north-west and south-east of Chalk River, it indicates the wind reversed on June 5, 2003 between 12:00 h and 18:00 h. In that way, Sheenboro, in the south-east of Chalk River (but 20 km north-east of Petawawa), shows a ^{133}Xe activity around 100 Bq m^{-3} between June 5, 2003 at 20:00 h and June 6, 2003 at 12:00 h. The NaI(Tl) measurement is as high as 400 Bq m^{-3} in Chapeau on June 5, 2003 at 21:00 h. Detections in Sheenboro and Chapeau occur after the wind reversal. The 15 December, 2003 event illustrates the variability of the wind and dispersion at the local scale.

High resolution simulations of the meteorological flow and ^{133}Xe transport were carried out with MM5 and FLEXPART modelling system (see Sections 5.2.1 and 5.2.2). As the location of the emission point is known (chosen as the Moly Stack), 1D temporal inverse problem was applied to the measurements issued by the Na(T1) detectors network in order to identify the emissions kinetics. The principle is the same as that described in Section 6.3.2.

Fig. 12 shows the calculated estimation of the amplitude of the radioxenon releases by Chalk River Laboratories. The results were obtained using MM5 wind fields (including a nudging towards NCEP/NOAA analysis, but no observational nudging), FLEXPART dispersion code, and Tikhonov regularization method. Comparable results were obtained with Backus–Gilbert regularization. Finally, the calculated radioxenon emissions were as follows:

- A limited first release of magnitude 1×10^{15} Bq took place between 03:00 h and 06:00 h on June 4, 2003. It was responsible for the detection in Deep River before the wind reversed.
- Minor releases of magnitude 1×10^{11}–7×10^{12} Bq took place between 06:00 h on June 4, 2003 and 09:00 h on June 5, 2003. They were responsible for detections in Deep River before the wind reversal and detections in Petawawa, Chapeau and Sheenboro after the wind reversal.
- A second release of magnitude 1×10^{15}–4×10^{15} Bq was emitted between 09:00 h and 12:00 h on June 5, 2003. It was not directly responsible for a detection as it occurred just before the wind started to reverse. However it reinforced the high detections in Petawawa and Chapeau when the plume was transported south-east down the valley of the Ottawa River accompanying the next release.
- A third release of magnitude 2×10^{15}–7×10^{15} Bq took place between 12:00 h and 15:00 h on June 5, 2003. As this release occurred during the wind reversal, the ^{133}Xe plume stagnated above the source region before it had been advected, with the contribution of the previous release, to the south-east. It was responsible for the high detections on Petawawa and Chapeau at respectively 20:00 h and 21:00 h on June 5, 2003, and, to a lesser extent, for the minor detections in Sheenboro not directly in the path of the plume.

In summary, the main calculated release occurred on June 5, 2003 between 12:00 h and 15:00 h in the interval of 1×10^{15}–7×10^{15} Bq. This release corresponds to the second hypothesis envisaged in the regional study (see Section 6.4.1).

6.5. Comparison of the results from the two models

6.5.1. The December 15, 2003 xenon event

The CMC model determined that the December 15, 2003 release had a source term of $\sim 10^{14}$ Bq. This is quite consistent with the CEA model which determined the source term to be between 1×10^{13} and 8×10^{13} Bq. Both models also performed well in predicting the temporal shape of the plume as it was detected down the Ottawa Valley.

The site-specific derived release limit (DRL) for Chalk River (Silke and De Waele, 2006) out of the stack on the Molybdenum production facility is 1.48×10^{15} Bq MeV per week for radioactive noble gases. A release of 10^{14} Bq for ^{133}Xe corresponds roughly to 8×10^{12} Bq MeV per event, or 4.8×10^{13} Bq MeV per week assuming 6 events per week, well below the DRL.

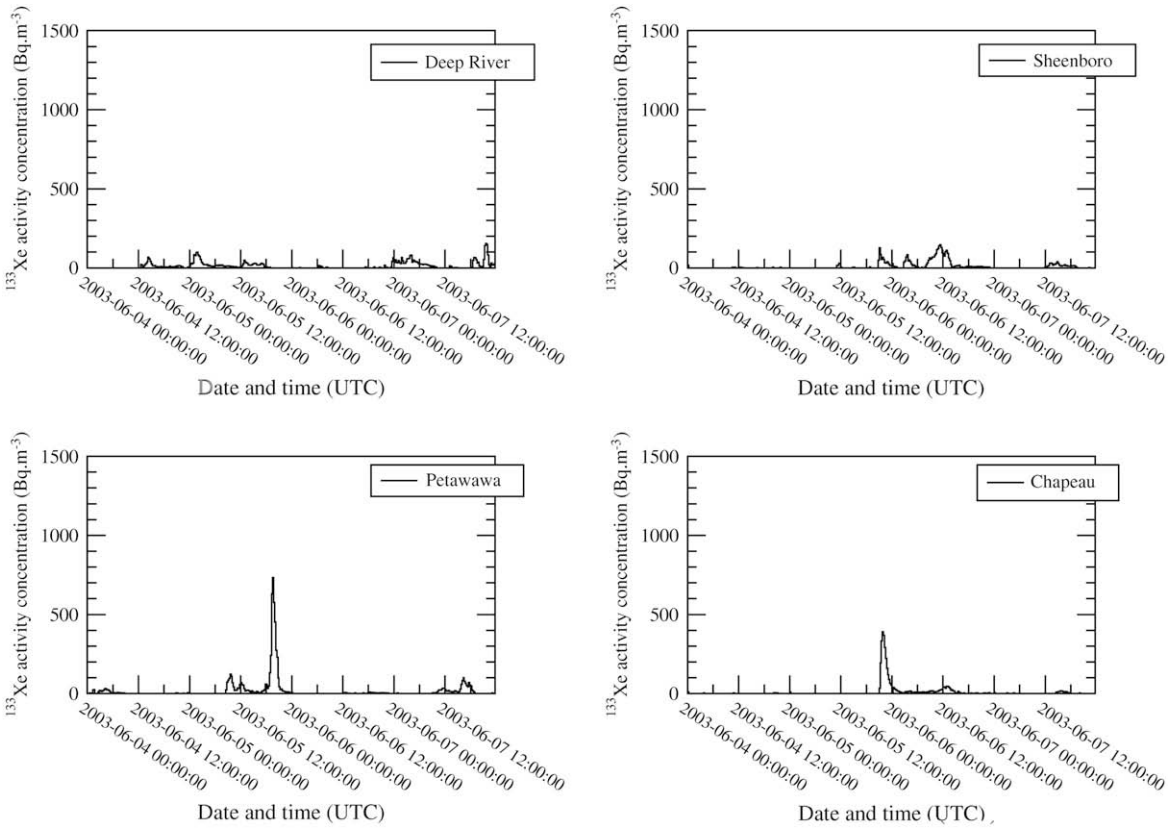

Fig. 11. Time histories of ^{133}Xe activity concentration measured by the NaI(Tl) detectors located down the valley of the Ottawa River. The time period is 00:00 h on June 4, 2003 to 00:00 h on June 8, 2003. There are seven NaI(Tl) detectors: Deep River, Sheenboro, Petawa, Chapeau, Westmeath, and Ottawa – RPB, but only the time histories of four of them with the highest signals are represented.

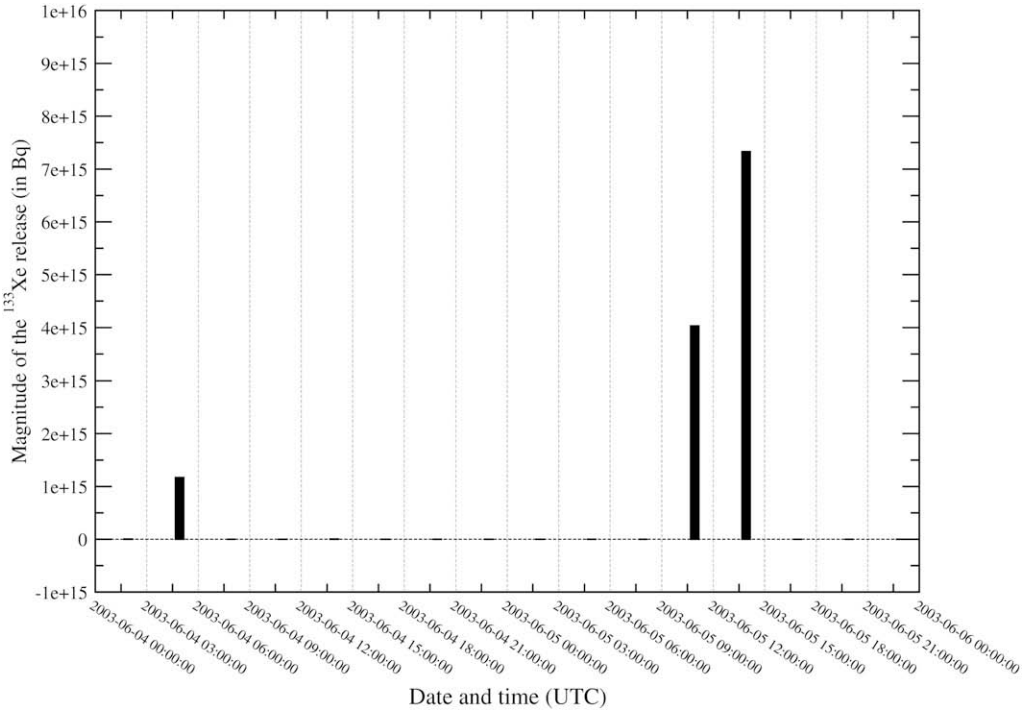

Fig. 12. Evolution with time of the magnitude (in Bq) of the releases by the Chalk River Laboratories. Simulations are based on MM5 wind fields (including a nudging towards NCEP/NOAA analyses, but no observational nudging), FLEXPART dispersion code and Tikhonov regularization method.

If we compare these source terms to the measured releases of Chalk River Laboratories, then we find the following. According to Silke and De Waele (2006), in 2003 CRL released 3.40×10^{13} Bq MeV per week of radioactive noble gases from the Moly facility. If we assume it was all ^{133}Xe, then the rate is 4.2×10^{14} Bq per week. If we then assume six equal releases, it is of the order of 7×10^{13} Bq, which is consistent with the estimate of both meteorological models.

6.5.2. The June 5, 2003 xenon event

The hypothesis that the June 5, 2003 event is a single release is not supported by the modelling of both organizations. In this hypothesis the models do not reproduce the NaI(Tl) observation near the source and the Ottawa measurement. However, according to the CMC modelling a release of the order of 10^{15} Bq could explain the large ^{133}Xe concentrations observed in Ottawa. This agrees very well with a release between 1×10^{15} and 7×10^{15} estimated by the CEA.

7. Conclusion

Health Canada has successfully installed and now operates a NaI(Tl) detector network in major population centres and around Canada's nuclear facilities. The primary goal of this network is to measure dose to the public, especially in the event of a nuclear incident or emergency. Given the detection sensitivity and the isotopic information provided by this network, it can also be used in support of scientific study. One sub-network of NaI(Tl) detectors in conjunction with operation of the SPALAX radioxenon analyser located in Ottawa has been used to verify meteorological transport models. These two systems have provided excellent environmental monitoring tools as they complement each other in terms of temporal resolution and activity concentration accuracy. A better understanding of the dispersion process in complex terrain was achieved by considering both model simulations and observational data. The application of meteorological modelling and detection ratios has been shown to be an excellent means of characterizing release sources.

The analysis of the radioxenon detections performed in the Ottawa Valley is of significant interest to support ongoing studies in atmospheric transport modelling and to provide a basis for model validation and intercomparison. In the future, other radioxenon detection events will be analyzed following the same principles as described in the paper.

The validation of the meteorological models' predicted source terms and measurements with actual releases from Chalk River demonstrates that the Ottawa Valley is an excellent "lab" to verify these models. The agreements and differences observed among the models provide a useful basis to identify best practices and approaches to model development and application. It is also a useful basis to understand their utility in emergency preparedness, source identification and support of noble gas studies for treaty verification.

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