Changes in radioxenon observations in Canada and Europe during medical isotope production facility shut down in 2008

Ian Hoffman · Kurt Ungar · Marc Bean · Jing Yi · René Servranckx ·
Calin Zaganescu · Nils Ek · Xavier Blanchard · Gilbert Le Petit ·
Guy Brachet · Pascal Achim · Thomas Taffary

Abstract Radioxenon concentration data collected at International Noble Gas Experiment (INGE) monitoring sites in the Northern Hemisphere are demonstrably impacted by emissions from medical isotope (MI) production facilities at Chalk River (CRL), Ontario and Fleurus, Belgium [Saey, J Environ Radioact 100(5):396–406, 2009]. Temporary cessation at these European MI facilities in the latter half of 2008 allowed an opportunity to assess the relative impact of distant MI facilities at INGE monitoring stations. In particular, the concentration distributions at the measurement sites and calculated sensitivities to the putative emission locations through atmospheric transport modelling (ATM) of meteorological Source Receptor Sensitivity (SRS) computations were used to study MI impacts.

I. Hoffman (✉) · K. Ungar · M. Bean · J. Yi
Radiation Protection Bureau of Health Canada,
775 Brookfield Road, Ottawa, ON K1A 1C1, Canada
e-mail: ian_hoffman@hc-sc.gc.ca

R. Servranckx · C. Zaganescu · N. Ek
Canadian Meteorological Centre of Environment Canada,
2121 Transcanada Highway, Dorval, QC H9P 1J3, Canada

X. Blanchard · G. Le Petit · G. Brachet · P. Achim · T. Taffary
Commissariat à l'Énergie Atomique, Bruyères-le-Châtel, 91297
Arpajon Cedex, France

Introduction

Several xenon isotopes, in particular ^{131m}Xe, ^{133}Xe, ^{133m}Xe and ^{135}Xe are important noble gases used in the monitoring of nuclear test explosions by member states of the CTBTO. However, large amounts of these Xe isotopes are also produced by the fission of uranium for the manufacture of medical isotopes such as ^{99}Mo, creating a hemispheric background signal that may mask the presence of a nuclear signal. While the individual isotopic components are emitted in different proportions from a nuclear explosion and a MI facility, there remains a substantial challenge in separating the industrial source component from any potential explosion component. Previous approaches to the problem of background emitters have looked at observations and the associated sensitivity of an observation site to the emitter location [1–5]. A logical extension to this approach that may assist in the background versus nuclear signal determination is to observe the same (correlated) signal at multiple detection sites at appropriate levels using particle dispersion atmospheric models.

Experimental

The combination of the CRL source location and the prevailing westerly wind direction means the noble gas monitoring stations in St. John's (Canada), and the Western Europe stations of Paris (France), Stockholm (Sweden) and Freiburg (Germany) would be the most suitable for study. Additionally, the European observation stations used are ideal because the transport time from CRL to all stations is typically similar. Having the same transport times means that the sampling and sensitivity dates to CRL are often aligned in time. Additionally, since all stations also have

some local sources of Xe emissions as shown by the presence of ^{135}Xe, we have some redundancy in that alternate stations are available when impacted by ^{135}Xe. Each sample collected has a sampling duration of 24 h except Stockholm. For the Stockholm observations—due to 12 h sampling period, the data collected was summed to make a 24 h sampling period so that it would match the other stations.

The ATM models used were Lagrangian dispersion models, where particles are released into a wind field at a source during the sample collection process. The models are run "backwards" in time, showing where the sampled air came from. SRS, or "sensitivity" in the context of this paper, is then a computation of the fractional number of particles in a geographic area during a time window in m^{-3}. For this paper, the differential time step of the ATM has been integrated from the standard model output of 3 h to 24 h to match the sampling cycles of the majority of observation stations. The ATM models used have a total (backward) resolution of 14 days prior to sample collection. Using the observations and the SRS values computed by Canadian Meteorological Centre's (CMC) ATM, a search for evidence of the first unambiguous multi-station observation of ^{133}Xe from a single source was performed.

Results and discussion

St. John's, Canada

^{133}Xe observed in St. John's is normally only due to emissions from CRL, and is typically on the order of a few mBq/m^3 to a few 10 s mBq/m^3. Previous observations from St. John's and the ATM provided by the CTBTO have demonstrated a good correspondence in both time and size with emissions from CRL and the calculated sensitivity of St. John's to CRL as shown in Fig. 1.

Figure 1, shows a long time series of observations and the SRS values associated with CRL. The SRS values, provided by the CTBTO, were computed using European Centre for Medium-Range Weather Forecasts (ECMWF) assimilated data, and were corrected for transit time decay. Throughout the entire time series there is only one day, labelled in the figure, where the station in St. John's was sensitive to Fleureus/Petten. The data presented in this figure is typical of the observations at St. John's under normal reactor operations at CRL.

Europe

First, we present in Fig. 2, a long time series of typical observation data from Paris when all MI facilities in Europe and Canada are operating, and on separate axes, data

from when the MI production in Europe had ceased and the only remaining large emitter in the hemisphere at CRL was operating. There is a clear distinction (difference in scale is approximately a factor of 10) between the normal operation of MI facilities and the shutdown period.

For the other European stations, a comparison of the Xe concentrations before and after MI shutdown period shows the Xe data to be very similar, which we do not present here. Data collected in the vicinity of Paris before and during the MI facility shut-down period in Europe in August through to early November in the vicinity of Paris is similar to the range of concentrations of observed in Freiburg, Germany and somewhat larger than the range of observations observed in Stockholm, Sweden. In all cases, there was a notable reduction of peak values of xenon observed by one to two orders of magnitude during the shut down period as is illustrated for the Paris data.

Estimation of CRL average daily emission rate and its impact on observations

CRL has an average daily emission of 17.3 ± 2.9 TBq, with a maximum of 52 TBq, and a minimum of 5 TBq [Phil Davies, Atomic Energy of Canada Limited (AECL), "Personal Communication"]. It typically has 2.3 ± 0.7 emission periods each day, each release having a duration of 2.6 ± 0.3 h, with 3.0 ± 0.3 TBq/h emitted during the release period [Phil Davies, Atomic Energy of Canada Limited (AECL), "Personal Communication"]. Observations from St. John's and the data above show a non-normal distribution of emissions; therefore the median rather than the mean is a more proper statistic when calculating the source strength.

Estimations of source strength based from the St John's measurements and models appear more self-consistent, perhaps not surprising as the observed concentrations and computed sensitivities were an order of magnitude higher than for the European sites. The median value of source term estimates from St John's measurements for CRL of on the order of 10^{13} Bq agrees relatively well with the limited knowledge available of CRL MI daily stack emissions, about 1.7×10^{13} Bq. Estimations from the European sites are an order of magnitude higher and sometimes two but not unreasonable considering the possibilities of CRL and local sources variations, transit times, and model limitations.

It was assumed that neither the average number of releases per day nor the size of each individual release changed during the observation period. Comparing the predicted impacts of CRL on noble gas observations in France, and in particular Paris shows that our reported observed ^{133}Xe values are reasonable and in agreement with previous studies [6]. Furthermore, it is also plausible that a similar impact would occur at the other European sites.

Fig. 1 Time Series of ^{133}Xe concentrations in St. John's NL and sensitivity to the Chalk River MI facility
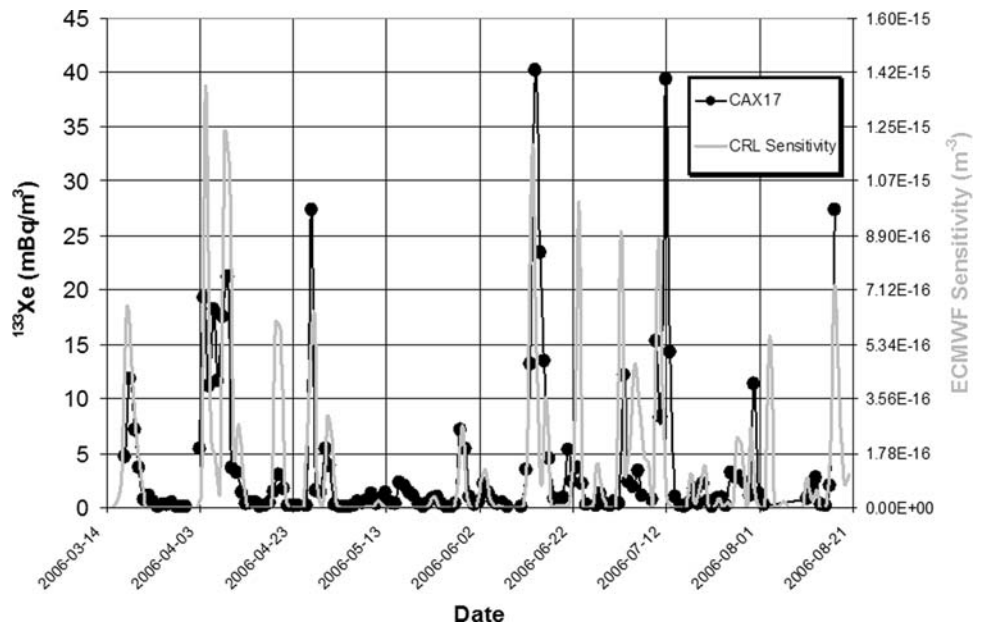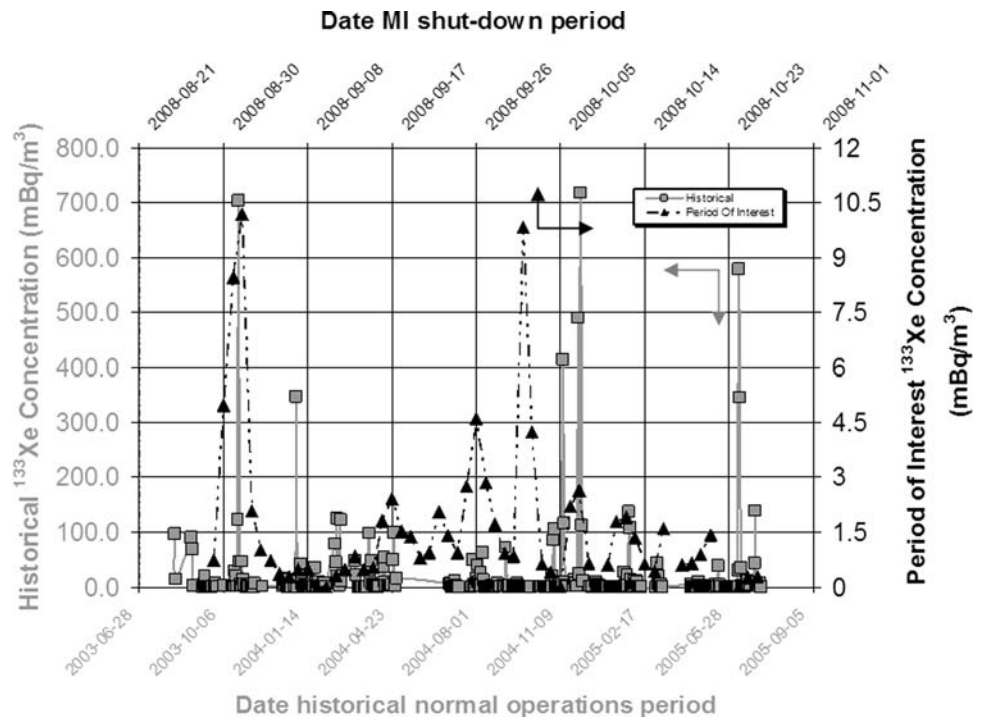

Fig. 2 Time Series of ^{133}Xe concentrations in the vicinity of Paris during recent European MI facility shut-downs and historical values during normal MI operations

Multi-station detection

To search for cases of a multi-station detection, we used a "temperature" greyscale graph as shown in Fig. 3. On the axes, the numbers of days from August 15, 2008 for sampling (*vertical axis*) and for sensitivity (*horizontal axis*) to CRL are shown for measurement sites in Canada and Europe. The stations are labelled as follows: Stockholm (SEX63), Freiburg (DEX33), Bruyères la Châtel (Paris),

and (CAX17). Each day of sensitivity (in m^{-3}) to CRL is represented by a greyscale square in the figure with the legend indicating the order of magnitude of sensitivity. The sensitivities shown were generated by Canadian Meteorological Centre, to precisely align with the noble gas sampling cycle. At St. John's, the typical transit time for an emission from CRL is roughly 2–5 days, while the corresponding European transit time is roughly 8–12 days. To approximately compensate for the additional dispersion in

transit to Europe and the associated decay, we multiply the sensitivity values for all of the European stations by a factor of 10. This "smoothing" allows the eye to see a similar temperature gradient across each station. Several potential cases of a multi-station detection are found by the presence of a vertical band of broadly similar "hot" colours. Vertical bands of similar colours represent similar sensitivities to releases from CRL and a matching observation at a station. The three candidate events for a multi-station observation of CRL are illustrated in the figure with an event number. The events selected range from being a potential 4-station detection to a 2-station detection.

Once the potential cases were identified, the data is then edited for cases where ^{135}Xe was present. The presence of ^{135}Xe with its short half life ($\tau_{1/2} = 9.14$ h) in a sample indicates the presence of a local source as the transit time does not allow a CRL source. Finally, we

integrate the daily sensitivities over the event period for which the station is solely sensitive to CRL. The sensitivity to CRL is plotted on a station by station basis in Fig. 4. The approximate timing of the events from Fig. 3, along with any observations of ^{135}Xe during the event windows is shown. While observations at St. John's appear uniquely related to releases from CRL, several observations at stations in Stockholm, Freiburg and Paris appear influenced by CRL, including some of the largest concentration observations of this time period. The events from the temperature gradient plot have been labelled with a rectangle showing the period of the event. Each data point inside each rectangle was used to calculate a CRL's ^{133}Xe source except when ^{135}Xe was present or when a sample was sensitive to CRL outside the defined event window. Table 1, summarizes the resulting events by presenting the medium source strength for each event

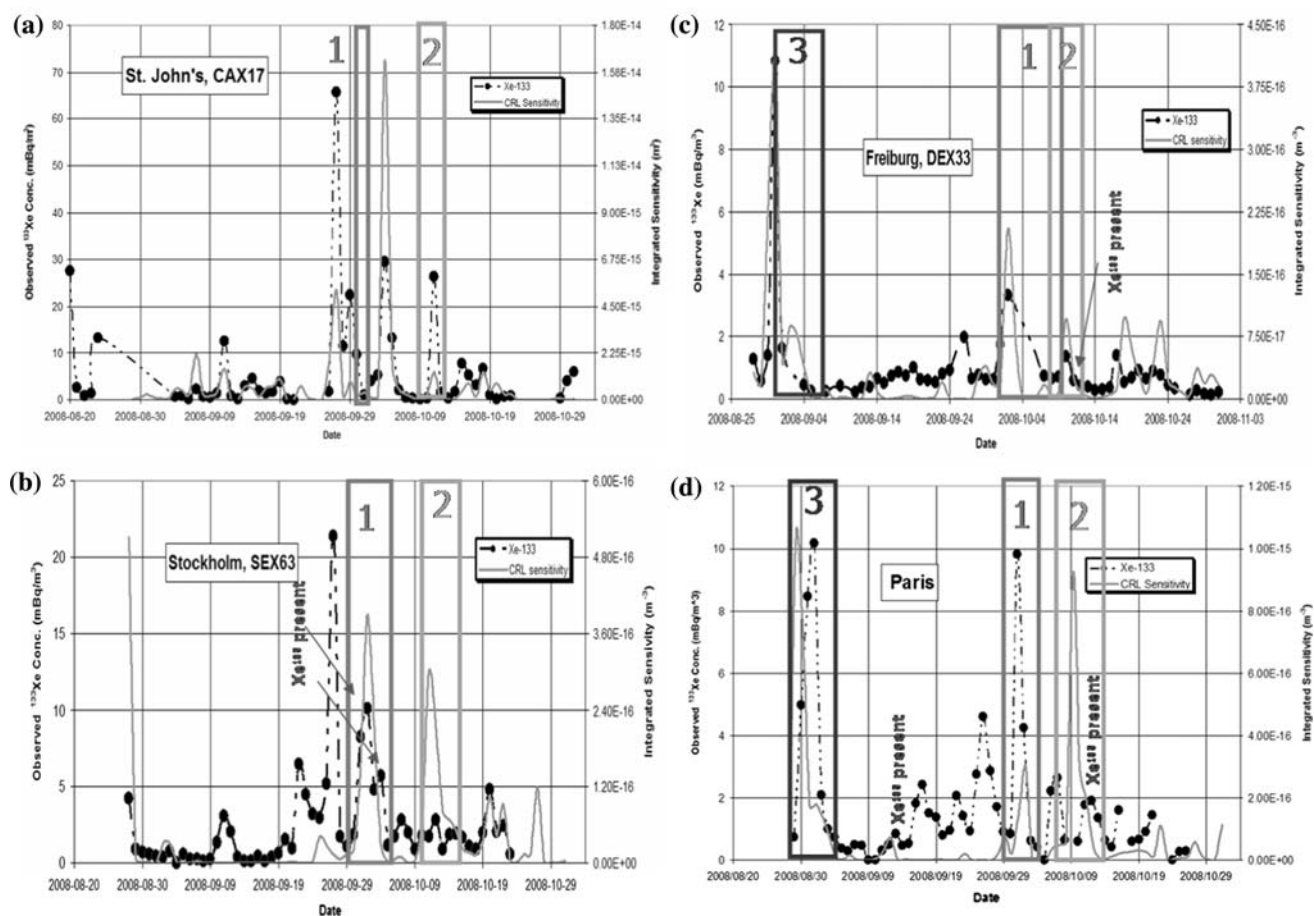**Fig. 3** Comparison of sampling date and dates of sensitivity to CRL
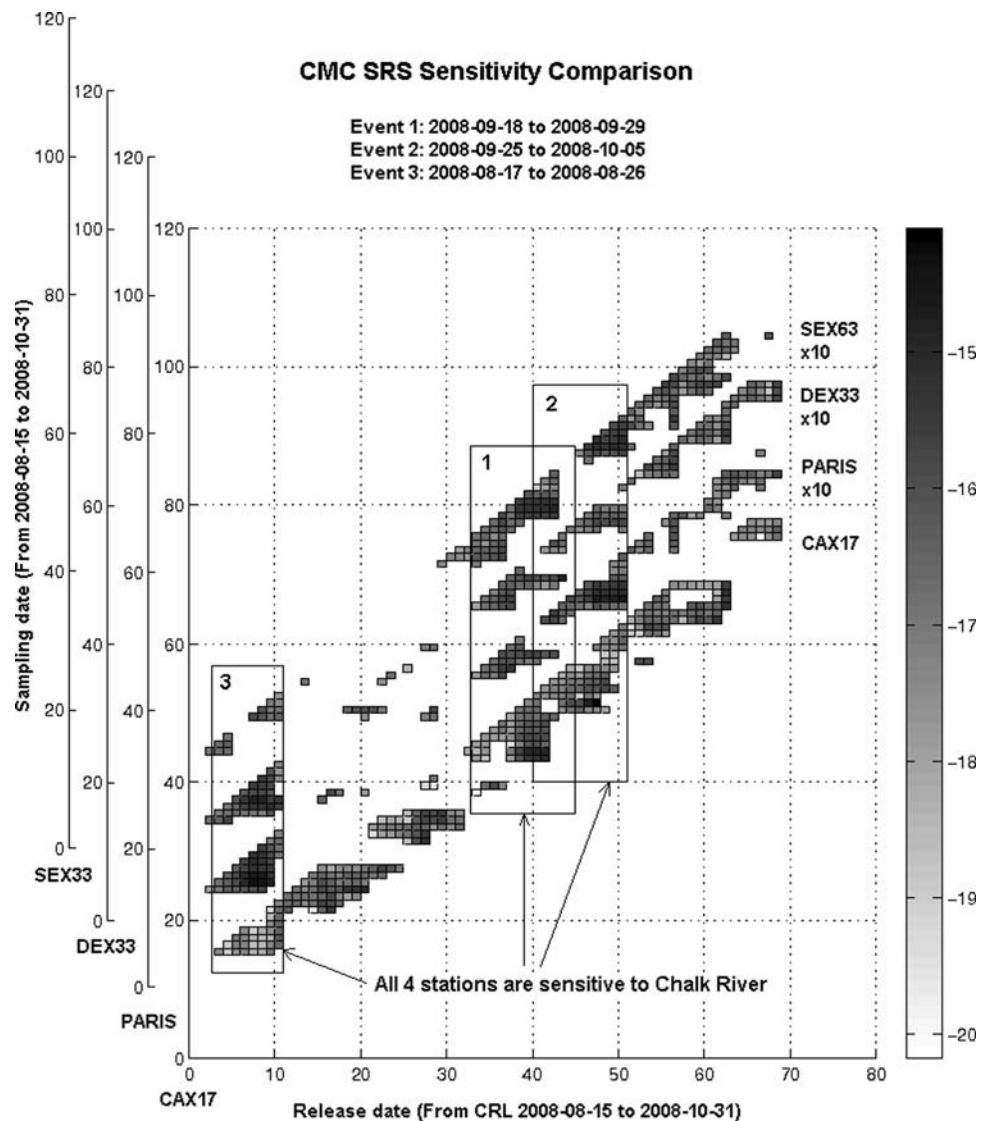
Fig. 4 Comparison of ^{133}Xe concentrations and integrated SRS (provided by the Canadian Meteorological Centre) at four measurement sites

Table 1 Implied source term and statistics of each event

Event	Station	Implied median source term (Bq/m^3)	N	High	Low
1	Paris	4.41E + 14	4	3.34E + 15	5.25E + 12
	SEX63	5.31E + 13	3	1.14E + 15	2.26E + 13
	DEX33	4.11E + 14	4	6.21E + 15	2.41E + 14
	CAX17	2.17E + 13	2	3.91E + 13	4.19E + 12
2	Paris	4.59E + 13	6	4.68E + 14	6.97E + 11
	SEX63	2.71E + 16	7	7.22E + 16	5.79E + 15
	DEX33	5.40E + 13	4	1.53E + 14	1.40E + 13
	CAX17	4.72E + 12	3	4.80E + 13	2.77E + 12
3	Paris	4.70E + 13	7	1.65E + 14	1.02E + 12
	DEX33	2.15E + 13	7	1.26E + 14	1.32E + 12

along with other relevant statistical information. In most cases the events agree on the source term within an order of magnitude amongst the stations, indicating a common CRL source assumption is reasonable. In the table, N represents the number of days of sensitivity that were used in the event time period, while the High and Low values are the maximum and minimum implied source term during the event.

Conclusion

The recent shutdown of medical isotope production facilities in Europe provided a special opportunity to estimate the impact of their radioxenon emissions on global and regional background levels of radioxenon in the Northern Hemisphere. The effect was profound, dropping observed levels of radioxenon in Europe by one to two orders of magnitude and revealing a residual influence of North American MI production contributing to radioxenon levels on the order of 1–10 mBq/m^3 at all three locations in Europe.

Detailed analysis of time series sensitivities among the four locations provided a means to interrelate specific days of observations at the four sites to common periods of release from CRL and similarities of measurement and sensitivity profiles, especially for the first event (Fig. 4). Actual estimates of release do not appear consistent with information about typical CRL stack releases except at the highest concentrations of xenon and calculated sensitivities observed in St. John's. Typically, the estimations for the European sites were in excess of an order of magnitude too high. Nevertheless, the ability to interrelate samples at different sites to a common location time and release suggests possible means of routine consideration of radioxenon observations on a network basis using SRS calculations.

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