

A long distance measurement of radioxenon in Yellowknife, Canada, in late October 2006

P. R. J. Saey,¹ M. Bean,² A. Becker,¹ J. Coyne,¹ R. d'Amours,³ L.-E. De Geer,¹ R. Hogue,³ T. J. Stocki,² R. K. Ungar,² and G. Wotawa¹

Received 7 May 2007; revised 17 August 2007; accepted 19 September 2007; published 16 October 2007.

[1] Between 21–25 October 2006, elevated levels of atmospheric xenon-133 were observed in Yellowknife (Canada). This station is located in an area where the background level of radioxenon is very low. The few measurements of xenon-133 above background in the last three years have been traced back to known nuclear facilities. The measurements in late October could not be linked to them. According to backward atmospheric transport models (ATM), the air that contained the measured radioxenon could have originated from the Korean Peninsula. On 9 October 2006, seismic networks world-wide recorded an event with characteristics of an underground explosion in the Democratic Peoples Republic of Korea. Forward ATM was performed using these coordinates. The results were consistent with the measurements in Yellowknife, more than 7000 km away. The order of magnitude of the amount measured is consistent with simple leak scenarios assumed for a low yield underground nuclear explosion on the Korean peninsula. **Citation:** Saey, P. R. J., M. Bean, A. Becker, J. Coyne, R. d'Amours, L.-E. De Geer, R. Hogue, T. J. Stocki, R. K. Ungar, and G. Wotawa (2007), A long distance measurement of radioxenon in Yellowknife, Canada, in late October 2006, *Geophys. Res. Lett.*, 34, L20802, doi:10.1029/2007GL030611.

1. Introduction

[2] Radioxenon measurements in Yellowknife are performed with a “Système de Prélèvement d'air Automatique en Ligne avec l'Analyse des radioxenon” (SPALAX) [Fontaine *et al.*, 2004], which samples and measures the air daily in Yellowknife, Canada. The SPALAX extracts xenon out of the air and measures the concentration activity of four isotopes/isomers, namely ^{131m}Xe , ^{133m}Xe , ^{133}Xe and ^{135}Xe by high purity germanium gamma ray spectroscopy [Stocki *et al.*, 2004]. The SPALAX is designed to maximize the efficiency of sampling and concentrating atmospheric radioxenon, to minimize the radon concentration in the process and to function continuously, automatically and reliably.

[3] In 1997 when the Provisional Technical Secretariat (PTS) of the Preparatory Commission (PrepCom) for the Comprehensive Nuclear-Test-Ban Treaty Organization

(CTBTO) [United Nations General Assembly, 1996] was established, no noble gas monitoring systems were commercially available that fulfilled the minimum requirements of the Treaty monitoring system, in particular a minimum detectable concentration (MDC) of 1 mBq/m³ for ^{133}Xe [Schulze *et al.*, 2000]. Therefore, the International Noble Gas Experiment (INGE) was established to design, develop, build, install and test such machines [Auer *et al.*, 2004]. The current equipment reaches MDC's of ^{133}Xe down to 0.1 mBq/m³. In October 2006, ten experimental noble gas stations were installed and delivered data to the PTS in Vienna, Austria. In October 2006 the active noble gas stations closest to DPRK were in Ulaanbaatar, Mongolia, at Spitsbergen, Norway, in Stockholm, Sweden and in Yellowknife, Canada. The relevant atmospheric circulation was generally from west to east disfavoured the first three sites while favouring the last, even though it is more than 7000 km away. Long-range transport of particulate radionuclides from Asia to this site has, however, been observed in the past [Wotawa *et al.*, 2006].

[4] A nuclear explosion creates fission products including radioactive noble gases in high yield. If the explosion is underground the enormous pressure generated can push these gases quickly into the atmosphere via cracks and fissures in the bedrock. Releases also occur during intentional access to the explosion cavity for investigative purposes. Finally, they can migrate through fractures to the surface driven by low pressure weather systems at a later time [Carrigan *et al.*, 1996]. It is reasonable to assume that a more immediate venting would inject a substantially larger fraction of the available noble gas into the atmosphere than a later seepage or venting during intentional access. No significant release of particulate radionuclides is expected in a well contained underground test. Therefore, noble gas (NG) monitoring is a sensitive technique for the detection of nuclear explosions underground or underwater [Perkins and Casey, 1996; Saey and De Geer, 2005]. Together with particulate radioactivity, noble gas monitoring is the only technique that has the potential to provide unmistakable evidence that an explosion is nuclear [De Geer, 1996].

2. Observations

[5] The experimental noble gas station in Yellowknife detected ^{133}Xe in excess of background on 21 October ($0.73 \pm 0.15(2\sigma)$ mBq/m³), and on 25 October ($0.49 \pm 0.14(2\sigma)$ mBq/m³) with somewhat lower values between 22 and 24 October (Figure 1b). Other xenon isotopes were not detected. The data were processed, analysed and reviewed at the PTS using in-house developed gamma spectral analysis software.

¹Provisional Technical Secretariat, Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organisation, Vienna, Austria.

²Radiation Protection Bureau, Health Canada, Ottawa, Ontario, Canada.

³Canadian Meteorological Centre, Meteorological Service of Canada, Environment Canada, Dorval, Quebec, Canada.

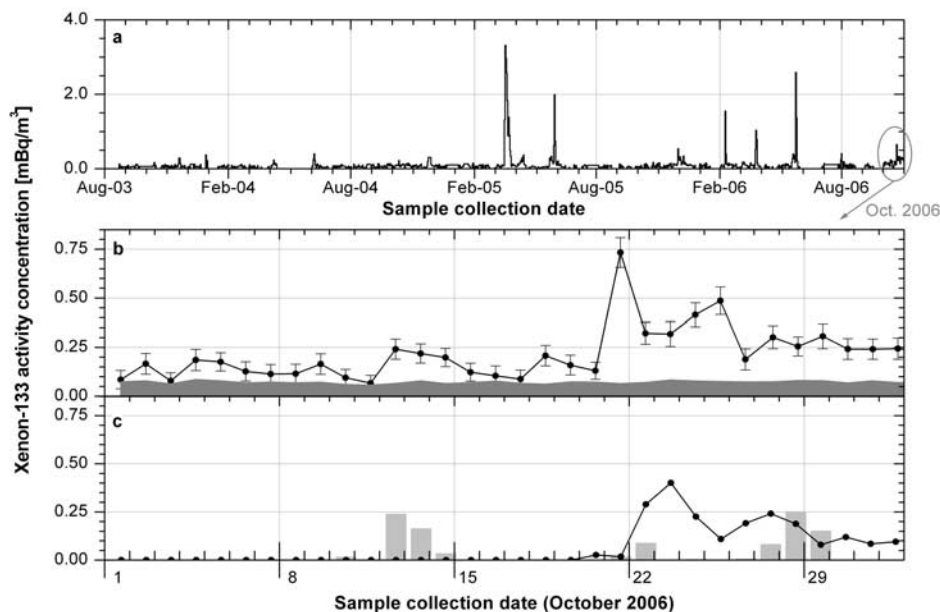


Figure 1. (a) The daily activity concentration of ^{133}Xe measured at Yellowknife, Canada, during three years from 15 August 2003 to 31 October 2006. (b) The observations of ^{133}Xe activity concentration at Yellowknife in October 2006 where the continuous grey pattern represents the minimum detectable concentration of ^{133}Xe . (c) The atmospheric transport modelling of the activity concentration of ^{133}Xe at the Yellowknife station, assuming the emission of 1 PBq of ^{133}Xe from the event on 9 October 2006. The small grey histogram in Figure 1c shows the contribution of the CRL facility when a maximum release rate is assumed for the whole month of October.

[6] The field measurements for samples ending October 24 and 25 were confirmed through re-measurement of a composite sample at the Health Canada's Radiation Protection Bureau (RPB) on a "Swedish Automatic Unit for Noble gas Acquisition" (SAUNA IIL) [Ringbom *et al.*, 2003] laboratory beta-gamma spectrometer.

3. Discussion

[7] On 3 October 2006, the DPRK televised an announcement that they would conduct a nuclear explosion. On Monday 9 October 2006 at 01:35 GMT, a seismic event of body wave magnitude 4.2 was estimated by the US Geological Survey to have occurred in north east DPRK, at 41.30°N, 129.10°E [Kim and Richards, 2007]. The German National Data Centre reported in Die Welt on 10 October 2006 that the event corresponds to an energy release of somewhat less than one kiloton explosion of TNT in hard rock [Bodderas, 2006]. The United States announced detection of "radioactive debris" [Office of the Director of National Intelligence, 2006] and the Republic of Korea reported detection of "xenon" (Yonhap News Agency, S. Korean gov't officially confirms N. Korea's nuclear test, Oct. 25, 2006) in the region shortly after the event, but no details have yet been published.

[8] Of the radioactive noble gases formed in a nuclear explosion, there are four xenon isotopes that could survive the time for transport and detection by noble gas monitoring systems: $^{131\text{m}}\text{Xe}$, $^{133\text{m}}\text{Xe}$, ^{133}Xe and ^{135}Xe , with half-lives of 11.934 days, 2.19 days, 5.243 days and 9.14 hours respectively (Evaluated Nuclear Structure Data File, Database version of July 2 2007, <http://www.nndc.bnl.gov/>

[ensdf/index.jsp](http://www.nndc.bnl.gov/)). The expected radioxenon signature is very sensitive to assumptions about the leakage process. For example, after a one kiloton plutonium explosion that was fully contained, ^{133}Xe would reach a maximum of about 10 PBq after 2.8 days. For initial considerations, we assume that 1 PBq of ^{133}Xe was emitted during the three hours following the explosion, as suggested by Perkins and Casey [1996] and Leith [2001]. Due to in-growth this corresponds to all ^{133}Xe available within the first few hours for a one kiloton explosion, or down to a fraction of about 10% within two days.

[9] To provide analysis of potential locations contributing to measurement data, the PTS has developed and operates an Atmospheric Transport Modelling (ATM) system [Wotawa *et al.*, 2003; Becker *et al.*, 2007]. Source-Receptor-Sensitivity (SRS) computations are routinely performed by the PTS. The Atmospheric Transport Model FLEXPART [Stohl *et al.*, 2005] is applied on analysed wind data provided by the European Centre for Medium-Range Weather Forecasts (ECMWF). The horizontal resolution of the data is 1° in longitude and latitude and the temporal resolution is 3 hours. The assumed maximum transport duration for the Yellowknife measurements was performed to cover the whole period after the event until 29 October. This SRS information was then utilised with a review tool to display "Fields of Regard" (FOR) for single samples (the catchment area related to one particular measurement). The SRS information was inverted to test specific area source assumptions in connection with the event and in connection with other possible sources for radioxenon measured in Yellowknife. Concentration time series resulting from the different source assumptions were compared with the measured time series.

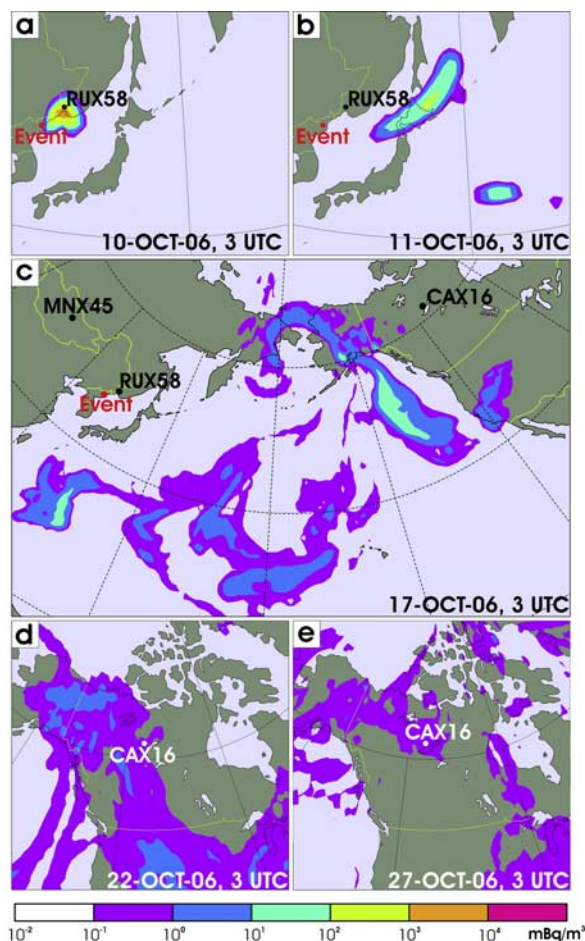


Figure 2. Calculated ground level concentrations of ^{133}Xe based on a 1 PBq emission at the time and coordinates of the 9 October DPRK explosion. The most interesting stages of the plume evolution are illustrated. (a) The initial dispersion from the event location (red dot) towards north-east reaching the scheduled PTS radio-xenon station RUX58 (Ussuriysk, Russian Federation). (b) The plume after two days when it passes Hokkaido, Japan. (c) After eight days, when its eastward evolution is decelerated by the mountain ranges of western North America. (d, e) The times when the plume is analysed to create the first and second peak at the PTS station CAX16 (Yellowknife, Canada). The station RUX58 was not installed at the time of the event.

[10] This system permits various data analyses, from a simple FOR to the reconstruction of complex measurement scenarios based on given source assumptions. Forward model computations from the event location were also performed using a 1 PBq release in the first 3 h after the event. The resultant plume is analysed to travel north-eastward towards Vladivostok, Russian Federation during the first 24 hours (Figure 2a) and across Hokkaido, Northern Japan one day later (Figure 2b). Due to vertical wind shear and uplifting the plume pattern soon develops a high level of complexity and a part of the plume passes Alaska and the Pacific Coast of North America on 17 October (Figure 2c). The many mountain ranges in Alaska, Yukon, and British Columbia are then a substantial obstacle so that it takes nearly another week before the Yellowknife area is

reached on 22 October (Figure 2d). On 27 October the plume is about to dilute and decay below the ^{133}Xe minimum detectable concentration of 0.1 mBq/m^3 (Figure 2e). As can be seen in Figure 1, the calculated time series at Yellowknife reproduces the measured signature, both the double peak structure and in quantity. There is a time lag of two days between the peaks in the model and in the observations, but it should be borne in mind that for a transport time in excess of two weeks, as in this case, a time deviation of two days is acceptable for the global dispersion model. According to the model, the station in Ussuriysk, Russian Federation (which was not installed at that time) would have measured a 100 fold stronger signal than the Yellowknife measurement, already two days after the event.

[11] A sensitivity analysis of the modelled measurement scenario regarding the release time was also conducted. The three-hour release was varied within the 72 hours following the event, in total 24 consecutive scenarios. These calculations showed that during the first 48 hours, any 3 hours release from the event location would have yielded the same double peak scenario as the one beginning immediately, although at smaller fractions of release. Even a release 72 hours later would not change the time of the peaks in the modelling results. The time lag to the observations is thus insensitive to the assumed timing of the leakage. It is rather to be attributed to the uncertainty of the dispersion model that is enhanced in particular during the second week when the plume travels across the mountainous terrain of western North America where only partly resolved vertical mixing processes begin to control the ground level arrival at Yellowknife. The therefore low sensitivity of the calculated time series in Yellowknife to the exact emission time during the first two days allowed for a more realistic scenario with only a fraction of the available ^{133}Xe leaking.

[12] Given the small but significant concentration of ^{133}Xe observed, detection of the other xenon isotopes at Yellowknife would not be expected from an explosion or common civil sources like nuclear power plants. Assuming that a maximum inventory of $^{131\text{m}}\text{Xe}$, $^{133\text{m}}\text{Xe}$ and ^{135}Xe from a 1 kT explosion leaked, the estimated concentrations at Yellowknife twelve days after the explosion would have been 0.1, 0.03 and $2 \times 10^{-8} \text{ mBq/m}^3$ respectively. These values are well below the detection limits of the SPALAX analyser for these isotopes of around 0.5 mBq/m^3 [Stocki et al., 2004].

[13] Other possible xenon sources for the Yellowknife readings were also considered. Three years of operational experience has shown that the station is located in an area where the background level of radioxenon from nuclear power plants (NPP) is very low [Saey et al., 2006]. Routine emissions from NPP regions in North America, Europe and Asia are not plausible sources of radioxenon excursions at Yellowknife as release rates at NPPs necessary to yield measurable activity in Yellowknife would be considered accidental in nature. There was no report of such occurrences during October 2006. Short periods of enhanced ^{133}Xe levels occurred occasionally during the last three years (Figure 1a), but each case could be traced back to emissions from a known medical radioisotope production facility, the Chalk River Laboratory (CRL) of Atomic Energy of the Canada Limited (AECL) 180 km west of from Ottawa, Canada. Hence, emissions from CRL merit careful consideration.

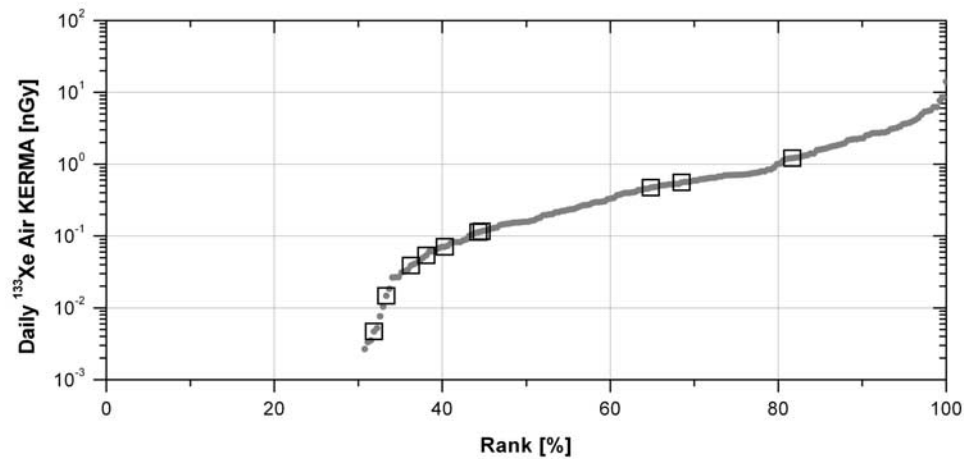


Figure 3. The frequency distribution of the dose rate in air KERMA, specifically due to ^{133}Xe for the year 2006, measured at typical NaI monitoring station Chapeau, near CRL. The x-axis indicates the fraction of data (rank) in %. The empty boxes indicate the measurements for the first 3 weeks of October 2006.

[14] Dosemetric estimates of radioxenon releases from the medical isotope facility at CRL were normal all through the month of October relative to long term history. These included the total radioxenon dose releases from the medical isotope production facility stack on a weekly basis, measured in TBq.MeV. It was independently corroborated with continuous monitoring data collected by RPB with NaI based spectroscopic monitors at seven stations in the vicinity of CRL, measured in nGy air KERMA. The weekly total xenon stack releases in the particularly relevant period to October 21 were essentially at the annual median value of 30 TBq.MeV [Thomson, 2003]. Radioxenon doses observed in the RPB operated monitoring network were typical of normal values in October ranging from 0.004 to 11 nGy air KERMA specifically due to ^{133}Xe (R. K. Ungar, unpublished data, 2006). Maximum values observed in this network for the month of October ranged from 5 to 25% of the maximum daily air dose observed in 2006, the ^{133}Xe value depending on the monitoring site (see Figure 3 for annual data and data of the first three weeks of October 2006).

[15] In addition, the daily ^{133}Xe emitted activity recorded by CRL from their radioisotopical production facility stack for the first 3 weeks in October were reported normal, these ranging from 5 to 44 TBq with a median value of 13.6 TBq of ^{133}Xe (B. Legree, Atomic Energy of Canada Limited, CRL, private communication, 2006). In the first half of October, Yellowknife measurements were actually sensitive to the CRL site providing an opportunity to scale the typical daily source strength of CRL emissions in October. Assuming that the ^{133}Xe observed in Yellowknife from 12 to 14 October and 27 to 29 October was exclusively due to CRL, a maximum CRL emission rate of 44 TBq/day can be assumed. Figure 1c gives the results of ATM calculations using the inversion technique of *Wotawa et al.* [2003] with a source term for all of October. It shows that little of the ^{133}Xe observed in Yellowknife from 21 to 25 October is likely caused by CRL emissions.

[16] This conclusion is supported by backward modelling for the Yellowknife measurements from 21 to 25 October confirming the sensitivity of the Yellowknife measurements to the Korean Peninsula. Figure 4 shows the Field of Regard

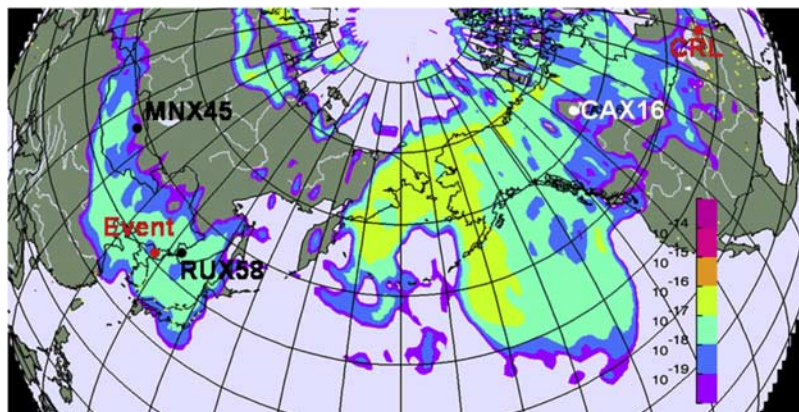


Figure 4. Field of Regard for the 21 October sample in Yellowknife referring to a ground level atmospheric injection of ^{133}Xe between 00 and 03 UTC on 9 October, 2006. The scale gives the dilution factor such that an emission of 1 PBq in, for example, a blue area is consistent with a detection in Yellowknife (CAX16) of 0.1–1 mBq/m³. This shows that Yellowknife is very sensitive towards East Asia including the DPRK, but not towards a potential alternate ^{133}Xe source, the Chalk River Laboratory in Ontario, Canada.

pertaining to the first peak collected at 21 October. It shows that for the three hours period comprising the DPRK event time the sensitivity to the CRL location is substantially less than that for the DPRK event location.

4. Conclusion

[17] Published seismic data confirmed that an event occurred on 9 October on the territory of the DPRK coinciding with the announced nuclear explosion. The event was estimated to be equivalent to the detonation of less than one kiloton TNT. Twelve days subsequent to this event, excess ^{133}Xe was detected at an experimental noble gas monitoring site in Yellowknife, Canada. Long range atmospheric backtracking and forward modelling of this noble gas revealed the signal to be not correspondent to any known release, but to be consistent with a release from the location and time of the DPRK event. The magnitude of the observed ^{133}Xe was consistent with a hypothetical 10% release of noble gas products estimated to be yielded by a one kiloton underground nuclear explosion. Finally the results demonstrate the continental scale remote ^{133}Xe detection capability for a low background station alike Yellowknife, Canada.

[18] **Acknowledgments.** The authors would like to thank the Director of the International Data Centre, Lassina Zerbo, for his continuous support in the presented work as well as the anonymous reviewers for their constructive comments. The views expressed in this publication are those of the authors and do not necessarily reflect the views of the Comprehensive Nuclear-Test-Ban Treaty Organisation Preparatory Commission or any of the participating institutions.

References

- Auer, M., et al. (2004), Intercomparison experiments of systems for the measurement of xenon radionuclides in the atmosphere, *Appl. Radiat. Isot.*, 60(6), 863–877.
- Becker, A., et al. (2007), Global backtracking of anthropogenic radionuclides by means of a receptor oriented ensemble dispersion modelling system in support of nuclear-test-ban treaty verification, *Atmos. Environ.*, 41, 4520–4534.
- Bodderas, E. (2006), War es wirklich die Bombe?, *Die Welt*, 10 Oct., 17.
- Carrigan, C. R., R. A. Heinle, G. B. Hudson, J. J. Nitao, and J. J. Zucca (1996), Trace gas emissions on geological faults as indicators of underground nuclear testing, *Nature*, 382, 528–531, doi:10.1038/382528a0.
- De Geer, L.-E. (1996), Atmospheric radionuclide monitoring: A Swedish perspective, in *Monitoring a Comprehensive Nuclear Test Ban Treaty*, edited by E. S. Huseby and A. M. Dainty, pp. 157–177, Kluwer Acad., Dordrecht, Netherlands.
- Fontaine, J. P., F. Pointurier, X. Blanchard, and T. Taffary (2004), Atmospheric xenon radioactive isotope monitoring, *J. Environ. Radiat.*, 72, 129–135.
- Kim, W.-Y., and P. Richards (2007), North Korean nuclear test: Seismic discrimination at low yield, *Eos Trans. AGU*, 88(14), 158.
- Leith, W. (2001), Geologic and engineering constraints on the feasibility of clandestine nuclear testing by decoupling in large underground cavities, *U.S. Geol. Surv. Open File Rep.*, 01-28.
- Office of the Director of National Intelligence (2006), Statement by the Office of the Director of National Intelligence on the North Korea nuclear test, *News Release 19-06*, Public Affairs Off., Washington, D. C.
- Perkins, R. W., and L. A. Casey (1996), Radioxenons: Their role in monitoring a comprehensive test ban treaty, *Rep. DOE/RL-96-1*, Pac. Northwest Natl. Lab., Richland, Wash.
- Ringbom, A., T. Larson, A. Axelsson, K. Elmgren, and C. Johansson (2003), SAUNA: A system for automatic sampling, processing and analysis of radioactive xenon, *Nucl. Instrum. Methods Phys. Res., Sect. A*, 508, 542–553.
- Saeey, P. R. J., and L. E. De Geer (2005), Notes on radioxenon measurements for CTBT verification purposes, *Appl. Radiat. Isot.*, 63, 765–773.
- Saeey, P. R. J., et al. (2006), Radioxenon background at high northern latitudes, *J. Geophys. Res.*, 111, D17306, doi:10.1029/2005JD007038.
- Schulze, J., M. Auer, and R. Werzi (2000), Low level radioactivity measurement in support of the CTBTO, *Appl. Radiat. Isot.*, 53, 23–30.
- Stocki, T. J., et al. (2004), Low level noble gas measurements in the field and laboratory in support of the Comprehensive Nuclear-Test-Ban Treaty, *Appl. Radiat. Isot.*, 61, 231–235.
- Stohl, A., C. Forster, A. Frank, P. Seibert, and G. Wotawa (2005), Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, *Atmos. Chem. Phys.*, 5, 2461–2474.
- Thomson, L. W. (2003), The Molybdenum-99 production facility annual safety review 2003, *Rep. AECL-MISC-303-03*, Chalk River Lab., Chalk River, Ont., Canada.
- United Nations General Assembly (1996), Follow-up to the advisory opinion of the International Court of Justice on the legality of the threat or use of nuclear weapons, *Resolut. 50/245*.
- Wotawa, G., et al. (2003), Atmospheric transport modelling in support of CTBT verification: Overview and basic concepts, *Atmos. Environ.*, 37(18), 2529–2537.
- Wotawa, G., L.-E. De Geer, A. Becker, R. D'Amours, M. Jean, R. Servranckx, and K. Ungar (2006), Inter- and intra-continental transport of radioactive cesium released by boreal forest fires, *Geophys. Res. Lett.*, 33, L12806, doi:10.1029/2006GL026206.
- M. Bean, T. J. Stocki, and R. K. Ungar, Radiation Protection Bureau, Health Canada, 775 Brookfield Road, A.L.6302D1, Ottawa, ON, Canada K1A 1C1.
- A. Becker, J. Coyne, L.-E. De Geer, P. R. J. Saeey, and G. Wotawa, Provisional Technical Secretariat, Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organisation, P.O. Box 1200, A-1400 Vienna, Austria. (paul.saeey@ctbto.org)
- R. d'Amours and R. Hogue, Canadian Meteorological Centre, Meteorological Service of Canada, Environment Canada, 2121 Trans-Canada Highway, Dorval, QC, Canada H9P 1J3.