Senate Standing Committee on Economics
ANSWERS TO QUESTIONS ON NOTICE
Innovation, Industry, Science and Research Portfolio
Supplementary Budget Estimates Hearing 2010-11
20 October 2010

AGENCY/DEPARTMENT: AUSTRALIAN NUCLEAR SCIENCE AND TECHNOLOGY ORGANISATION

TOPIC: Xenon Emissions

REFERENCE: Question on Notice (Hansard, 20 October 2010, E4)

QUESTION No.: SI-1

Senator LUDLAM—Okay. I have got a couple of issues I would like to raise. I have not raised with ANSTO before the issue reported in the press about a release of radioactive xenon-133 from the facility when I think it was being commissioned. I just wonder if you could provide us with a quick background of that event, and particularly for some of the material that ANSTO provided—and thank you for providing some information so quickly on receipt of my letter. You provided a table of monthly discharges of radioxenon between November 2008 and February 2009. What I am seeking would be something more like real-time monitoring reports that would actually show us that spike, rather than something smoothed out across a whole month.
Dr Storr—That information is available on ANSTO's website. If you go to the website you will be able to find real-time monitoring data for discharges.
Senator LUDLAM—Okay, so if we go to the dates when it was alleged in the newspapers that those releases occurred, that will actually show us the xenon spike that was detected in Melbourne?
Dr Storr—I will have to check that it goes back that far.
Senator LUDLAM—Well, it is not that long ago—only two years.
Dr Storr—I will take it on notice.
Senator LUDLAM—Okay. If it is not there could you undertake to provide us with that please?

ANSWER

Real-time Environmental Radiation Monitoring from a monitoring station located in Engadine, 3 km south-east of Lucas Heights in New South Wales, has been available on ANSTO's website since November 2007. The link to the Radiation Monitoring page is as follows: http://www.ansto.gov.au/discovering_ansto/what_does_ansto_do/live_weather_and_pollution_data/environment_radiation_monitoring The information on the page is updated at half hour intervals and at this stage the page does not display historical data.

Real-time monitoring is not available from the International Monitoring Station in Melbourne, which is operated by the Australian Radiation Protection and Nuclear Safety Agency (ARPANSA). However, historical data on the releases of xenon are publically available in an article in the Journal of Environmental Radioactivity 101 (2010) pages 353-361, which was co-authored by ARPANSA, ANSTO and their counterparts at the Vienna University of Technology. A copy of the article titled *Evaluation of Radioxenon releases in Australia using Atmospheric Dispersion Modelling Tools* is attached to this response (Attachment A). A graph showing the spikes in the detection of the xenon is on page 359 of the article.

ATTACHMENT A

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Evaluation of radioxenon releases in Australia using atmospheric dispersion modelling tools

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ABSTRACT

The origin of a series of atmospheric radioxenon events detected at the Comprehensive Test Ban Treaty Organisation (CTBTO) International Monitoring System site in Melbourne, Australia, between November 2008 and February 2009 was investigated. Backward tracking analyses indicated that the events were consistent with releases associated with hot commission testing of the Australian Nuclear Science Technology Organisation (ANSTO) radiopharmaceutical production facility in Sydney, Australia. Forward dispersion analyses were used to estimate release magnitudes and transport times. The estimated ^{133}Xe release magnitude of the largest event (between 0.2 and 34 TBq over a 2 d window), was in close agreement with the stack emission releases estimated by the facility for this time period (between 0.5 and 2 TBq). Modelling of irradiation conditions and theoretical radioxenon emission rates were undertaken and provided further evidence that the Melbourne detections originated from this radiopharmaceutical production facility. These findings do not have public health implications. This is the first comprehensive study of atmospheric radioxenon measurements and releases in Australia.

1. Introduction

Many nuclear-related processes can result in the release of radioxenon isotopes to the atmosphere. Such processes, which vary considerably in their public health or environmental significance, include: the production and use of radiopharmaceuticals; nuclear and research reactor operation and maintenance; nuclear fuel processing plants; above- or below-ground nuclear detonations; and other processes resulting in the fission of nuclear material. In order for a detected radioxenon event to be unequivocally attributed to a suspected nuclear detonation a detailed understanding of the behaviour and magnitude of releases from other potential sources is required.

Xenon isotopes are produced with a relatively high yield in nuclear explosions because their mass is near the maximum of the mass distribution of ^{235}U and ^{239}Pu fission products. The four key xenon isotopes with half lives sufficiently long for them to be detected after atmospheric transport on synoptic timescales are: ^{131m}Xe (11.84 d), ^{133}Xe (5.24 d), ^{133m}Xe (2.19 d), and ^{135}Xe (9.14 h). In nuclear explosions these isotopes are produced in large activities (e.g. 10^{16} Bq for a 1 kton atmospheric nuclear explosion), with unique activity ratios (Saey, 2009).

Xenon gases are highly volatile and, being inert, they are not susceptible to wet or dry atmospheric removal mechanisms. Consequently, once released to the atmosphere they are simply transported down-wind while radioactively decaying away. These characteristics make xenon gases ideal tracers of nuclear-related processes. For example, in 2006 back trajectory analyses were employed to link elevated levels of ^{133}Xe observed in Yellowknife, Canada, to a low yield underground nuclear explosion on the Korean Peninsula more than 7000 km away (Becker et al., 2007; Saey et al., 2007).

With half lives that vary from hours to weeks, some radioxenon isotopes can be transported great distances, leading to spatially and temporally heterogeneous background activity concentrations and activity ratios at the hemispheric scale. Typical mean ^{133}Xe values in the industrialised nations of the northern hemisphere are between 0.2 and 6 mBq m^{-3}, with occasional detection of the less abundant ^{131m}Xe, ^{135}Xe and ^{133m}Xe (Saey, 2009). In closer proximity to nuclear and radiopharmaceutical isotope production facilities all four isotopes can reach levels up to a few thousand mBq m^{-3} (Saey, 2009; Kalinowski et al., 2005; UNSCEAR, 2000).

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In Australia, the largest source of atmospheric background radioxenon is from the irradiation of 19.75% Low Enriched Uranium (LEU) (NSRB, 2009) to produce ^{99}Mo at the nuclear facilities of the Australian Nuclear Science and Technology Organisation (ANSTO), in southern Sydney. ANSTO operates Australia's only nuclear reactor, the Open Pool Australian Light-water reactor (OPAL) for research and radiopharmaceutical isotope production. Radiopharmaceutical isotope production was started again at the end of 2008 after the opening of a new facility. Australia also imports small clinical amounts of ^{133}Xe for in-vivo lung ventilation and image studies. Other potential contributors to background radioxenon activities include regional nuclear and radiopharmaceutical production facilities in the Southern Hemisphere. Nuclear Technology Products (NTP) Radioisotopes in South Africa is considered the fourth largest producer of ^{99}Mo, supplying 10% to the world market (NSRB, 2009). Comisión Nacional de Energía Atómica (CNEA) in Argentina supplies ^{99}Mo for its domestic market and to some South American countries (NSRB, 2009). With its limited number of potential source terms for radioxenon emission, only 21 nuclear power plants and research reactors are located in the southern hemisphere compared to hundreds in the northern hemisphere (IAEA, 2010), and the limited potential for mixing between hemispheres (UNSCEAR, 2000). The southern hemisphere can provide a unique low background radioxenon environment to study the behaviour and origins of single releases.

Due to the pervasive nature of radioxenon isotopes, long-term, ground-based monitoring is required to characterise the typical variability in regional background activities. Using established background activities as a reference it is easier to quantitatively distinguish local from remote and expected from unexpected releases, and thus better gauge their potential public health or environmental consequences. When analysed in conjunction with near-real-time weather forecasts and atmospheric transport models, radioxenon isotope detection data can also be used to make predictions of source locations and release magnitudes. To these ends the International Monitoring System (IMS) was established, which constitutes the verification network of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). The Australian Radiation Protection and Nuclear Safety Agency (ARPANSA) operates and maintains two automated low level radioxenon monitoring stations as part of the IMS, located in Melbourne and Darwin.

This study, which focuses on detections by the Melbourne IMS station (37°42′16″S; 145°6′1″E at approximately 10 m above ground level) between November 2008 and January 2009, constitutes the first characterisation of radioxenon background in Australia. A number of ^{133}Xe detections were made over this period, as well as detections of ^{133m}Xe and ^{135}Xe. In conjunction with these observations, the backwards and forwards tracking analysis capabilities of selected atmospheric transport models are used to hypothesise likely source location and magnitudes. Stack monitoring results from the radiopharmaceutical production facility pertaining to the timing and magnitude of releases enabled subsequent verification of these hypotheses.

2. Detection of radioxenon isotopes

Atmospheric radioactive xenon gas was detected using the fully automatic SAUNA II (Swedish Automatic Unit for Noble gas Acquisition) system (Ringbom et al., 2003). The sampling system consists of two parallel sampling ovens. Moisture and carbon dioxide is removed from the sampled air using thermoelectric coolers and molecular sieves. The air then enters charcoal traps where the xenon is absorbed. Each sampling oven samples air on alternating 6 h cycles. Once the first 6 h sample is collected it is absorbed on a charcoal trap in the processing oven. Once the second 6 h sample is collected it is combined with the first sample in the processing oven. The combined sample then undergoes preparative gas chromatography for further purification and separation of radon. The sample volume is quantified by measuring the stable atmospheric xenon using a thermal conductivity detector. The concentrations of the isotopes ^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe are measured using a beta-gamma coincidence technique; the counting time is approx. 12 h. This technique reduces the ambient background, and the recording of the beta signal allows for high sensitivity measurements of the metastable isomers. Average minimum detectable concentrations (MDC) are well below 1 mBq m^{-3} for all four isotopes. Individual variations in the MDC occur for each sample.

A series of radioxenon detections were made at the Melbourne IMS station between November 2008 and January 2009 (Table 1). One of the most significant of these occurred from November 25 to 27. A maximum ^{133}Xe activity concentration of 6.2 ± 0.2 mBq m^{-3} occurred on 26 November 2008. The spectrum of this sample is shown in Fig. 1. The γ coincidence spectrum shows clearly the 30 keV X-ray peak and the 81 keV gamma peak of ^{133}Xe. Other detection events ranged from 0.4 to 2.4 mBq m^{-3} for ^{133}Xe.

All times and dates in this paper are in UTC unless otherwise specified.

3. Description of atmospheric transport models and meteorological data

Based on high resolution meteorological data and using atmospheric transport models, it is possible to trace the various atmospheric pathways of any radionuclide detected at a monitoring station back to the area where it may have originated. Three numerical tracking models (FLEXPART within WEB-GRAPE, HYSPLIT and RIMUFF within ARGOS) were applied to the situation, with each model applied in a combination of backward and/or forward analysis scenarios.

Evaluation of the model outputs were limited by the difficulty in assigning uncertainties to scenarios. These uncertainties stem from predicting spatial and temporal meteorological conditions from complex air movements over large distances. For example, a small deviation from a predicted to a real wind direction can cause significant differences in the activity concentration levels at the detection point. Approximation of source term activities and their rate and time of release are also significant contributors to uncertainty. The variability in estimating these terms were incorporated in the scenarios by applying a variable numerical approach such as sequentially increasing release times. This enabled estimation of the possible source region and release activity magnitude (or range) that would best match the radioxenon detections.

Table 1
Radioxenon measurements at the Melbourne IMS station detected from November 2008 and February 2009.

Collection stop date and time (12 h collection)	^{133}Xe activity concentration (mBq m^{-3})	^{133m}Xe activity concentration (mBq m^{-3})	^{135}Xe activity concentration (mBq m^{-3})
26 November 2008 07:38 h	3.0 ± 0.1	<0.25	<0.44
26 November 2008 19:38 h	6.2 ± 0.2	0.30 ± 0.11	<0.48
27 November 2008 07:38 h	1.7 ± 0.1	<0.20	<0.50
12 December 2008 07:38 h	2.0 ± 0.1	0.14 ± 0.07	0.24 ± 0.14
12 December 2008 19:38 h	1.6 ± 0.1	0.10 ± 0.06	0.25 ± 0.14
13 December 2008 07:38 h	0.4 ± 0.1	<0.13	<0.48
21 December 2008 07:38 h	2.4 ± 0.1	<0.22	0.33 ± 0.15
21 December 2008 19:38 h	1.2 ± 0.1	0.14 ± 0.06	<0.49
28 January 2009 19:38 h	0.8 ± 0.1	<0.16	<0.56
29 January 2009 07:38 h	0.5 ± 0.1	<0.14	<0.46

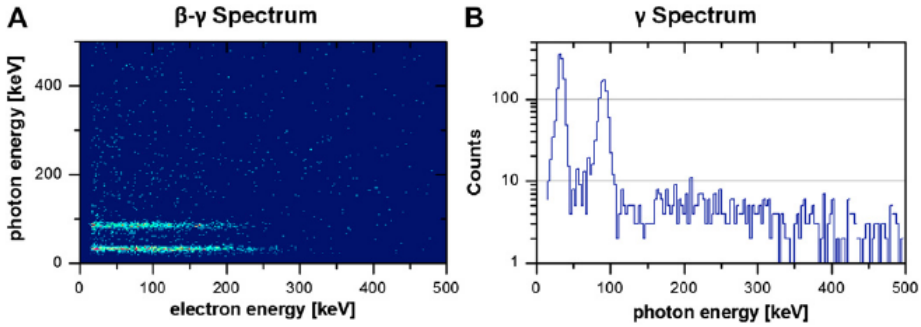

Fig. 1. The beta-gamma coincidence spectrum (A) and its gamma projection (B) for the sample collected between 26 November 2008 07:38 and 19:38 h.

3.1. FLEXPART within WEB-GRAPE

WEB-GRAPE stands for WEB connected GRAPhics Engine and is used to support the verification of the CTBT. WEB-GRAPE uses the diagnostic Lagrangian particle model FLEXPART (Stohl et al., 1998, 2005) to perform atmospheric dispersion modelling simulations. Using FLEXPART, WEB-GRAPE can correlate radioxenon measurements with resulting signals from possible source scenarios on a global scale to identify potential source locations of the detected radionuclides (Becker and Wotawa, 2007). The model outputs are integrated backward in time based on global weather prediction data to yield a concentration-dilution map of possible source locations. It is limited to dispersion at the surface level (0–30 m) on a global grid of 1° by 1° latitude-longitude intersections at 3 h intervals up to 14 d backwards tracking. The software receives as input numerical weather prediction data from two world-leading centres (European Centre for Medium-Range Weather Forecasts and US Centre for Environmental Prediction) in near-real-time. Terrain height is defined within the numerical weather prediction data. This study used Version 1.3 of WEB-GRAPE operating on a PC platform with a Windows XP operating system connected to the secure CTBT database.

3.2. HYSPLIT

The HYSPLIT, HYbrid Single-Particle Lagrangian Integrated Trajectory, model is designed to support a wide range of simulations related to the long-range transport, dispersion, and deposition of pollutants (Draxler, and Hess, 1998; Draxler and Rolph, 2003; Rolph, 2003; Draxler, 2007). The model calculation method is a hybrid between Eulerian and Lagrangian approaches. Advection and diffusion calculations are made in a Lagrangian framework while concentrations are calculated on a separate grid defined by 0.3° latitude-longitude resolution. Archived meteorological datasets were used in Global Data Assimilation System (GDAS) format. Terrain height is defined within the meteorological data. The model was used for backward trajectory and forward dispersion analysis, with no wet or dry deposition. This study used the online version 4 of HYSPLIT (http://www.arl.noaa.gov/ready.html) accessed between January and June 2009.

3.3. RIMPUFF within ARGOS

ARGOS (Accident Reporting and Guidance Operational System) is a decision support tool applied to chemical, biological, radiological and nuclear emergencies (PDC, 2008). Short and medium range atmospheric dispersion modelling is conducted using the RIMPUFF (Riso Meso-scale Puff) model within ARGOS (Mikkelsen et al., 2007). RIMPUFF simulates airborne materials by sequentially releasing a series of Gaussian shaped puffs at a fixed rate (Thykier-Nielsen et al., 2005). The RIMPUFF model produces forward time predictions of material dispersion and deposition (forward dispersion analysis) and is capable of using local numerical weather prediction data. The maximum spatial grid resolution is 50 m. The RIMPUFF model has previously been used in evaluation studies at the ANSTO site (Williams et al., 2005). RIMPUFF has been applied to the radioxenon release using Limited Area Prediction System (LAPS) numerical weather data provided by the Melbourne Bureau of Meteorology. The LAPS data covers all of Australia and some of Asia, at a resolution of 37.5 km with 61 terrain following vertical levels (Puri et al., 1998; BOM, 2007). Output data is in 3 h intervals and is assessed every 12 h. This study used version 8.064 of the RIMPUFF model within version 8.3.8.0 of the ARGOS client, operating on PC platform with Microsoft XP.

4. Initial assessment of potential source release locations

In order to determine the likely source of radioxenon detected at the Melbourne IMS station, a range of backwards tracking assessments were performed using WEB-GRAPE for the 26 November detection. Backwards tracking was based on the largest ^{133}Xe detection event in Melbourne, for various detection times over the single day from 26 November 0:00 h to 27 November 0:00 h.

Long range backwards tracking results covered a period of up to 14 d, providing coverage over most of the Southern Hemisphere that included the two largest international producers of ^{99}Mo in the region, the most likely sources for ^{133}Xe release (Fig. 2 A). Backwards tracking predicts a travel time in the range of 9–14 d from the NTP Radioisotopes in South Africa and the CNEA facility in Argentina. The resulting dilution factor is approximately 10^{-18}, which corresponds to a source release in the PBq range to produce the required detection in Melbourne. Both scenarios are unrealistic, considering the average annual release for the NTP facility is 4.1 PBq from over 300 days of operation per year (Saey, 2009). It should be noted that there were no reported nuclear weapon detonations during the time frame examined in this study.

Short range backwards tracking covered a period of up to 3 d. Results indicated a strong possibility that the release was initiated at the radiopharmaceutical facility of ANSTO, Lucas Heights. Fig. 2B displays a concentration-dilution map at 25 November 2008 9:00 h for a collection period ending 26 November 2008. A travel time to Melbourne of approximately 36 h is predicted with a dilution factor of 10^{-15}. The release source term is estimated to be in the range of 1–10 TBq. This compares well to radiopharmaceutical facilities in Canada, Belgium and South Africa. At these facilities the average daily releases are in the range of 4.6–16 TBq (Saey, 2009). Fig. 2C

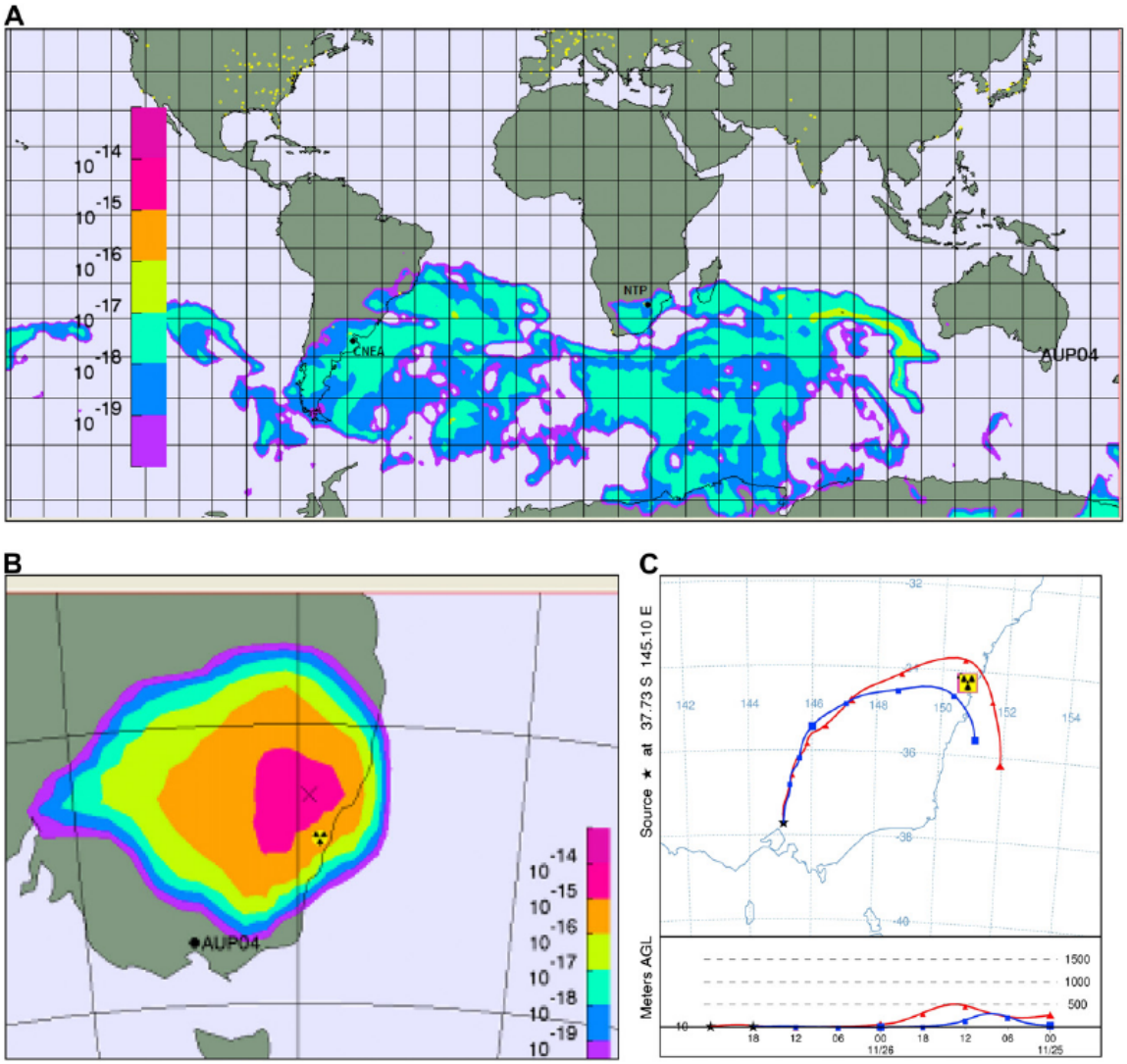

Fig. 2. Backtracking snapshots based on the largest ^{133}Xe detection event detected by the Melbourne IMS station for the collection period ending 26 November 2008. AUP04 denotes the location of the Melbourne IMS station. (A) WEB-GRAPE dilution map for 13 November 15:00 h (B) WEB-GRAPE dilution map for 25 November 2008 9:00 h (C) HYSPLIT backwards trajectory output starting 26 November 18:00 h and 27 November 2008 0:00 h.

displays the HYSPLIT backward trajectory analysis, showing two likely pathways back to the ANSTO region (34°03′06″S; 150°58′41″E) for a time series of 26 November 18:00 h and 27 November 2008 0:00 h.

5. Detailed assessment of source release

Forward atmospheric transport modelling can predict where radionuclides may travel from their known point of release and allows the theoretical magnitude of the release to be estimated. Forward dispersion analysis was performed using the RIMPUFF model within ARGOS. Releases from ANSTO were simulated using a 10 TBq release of ^{133}Xe over 1 h, in 10 min puffs with a spatial grid resolution of 5 km. LAPS weather data was used and a release stack height of 20 m was assumed. An hourly release was chosen to give good temporal resolution, with 10 min puffs to simulate the possibility of multiple venting over the release. Releases were simulated from the 23 to 25 November and dispersion analysis was performed until the end of the third detection period (27 November 8:00 h) in Table 1.

The predicted release periods of plumes that passed over the Melbourne IMS station within the detection periods ranged from 24 November 6:30 h to 25 November 9:30 h. Within this range, multiple source releases are possible; two examples are shown in Fig. 3A and B.

HYSPLIT forward dispersion analysis was applied in concentration mode at a release height between 10 and 50 m for release duration of 1 h with no dry or wet deposition. Simulations over a similar time period to RIMPUFF showed the plume would pass over the Melbourne IMS station 24–36 h after release, shown in Fig. 3C and D. Transport times for these durations fall within detection periods and the plume direction is consistent with the RIMPUFF modelling.

The predicted ^{133}Xe activity concentrations for plumes passing over the Melbourne IMS station for each of the first three detection

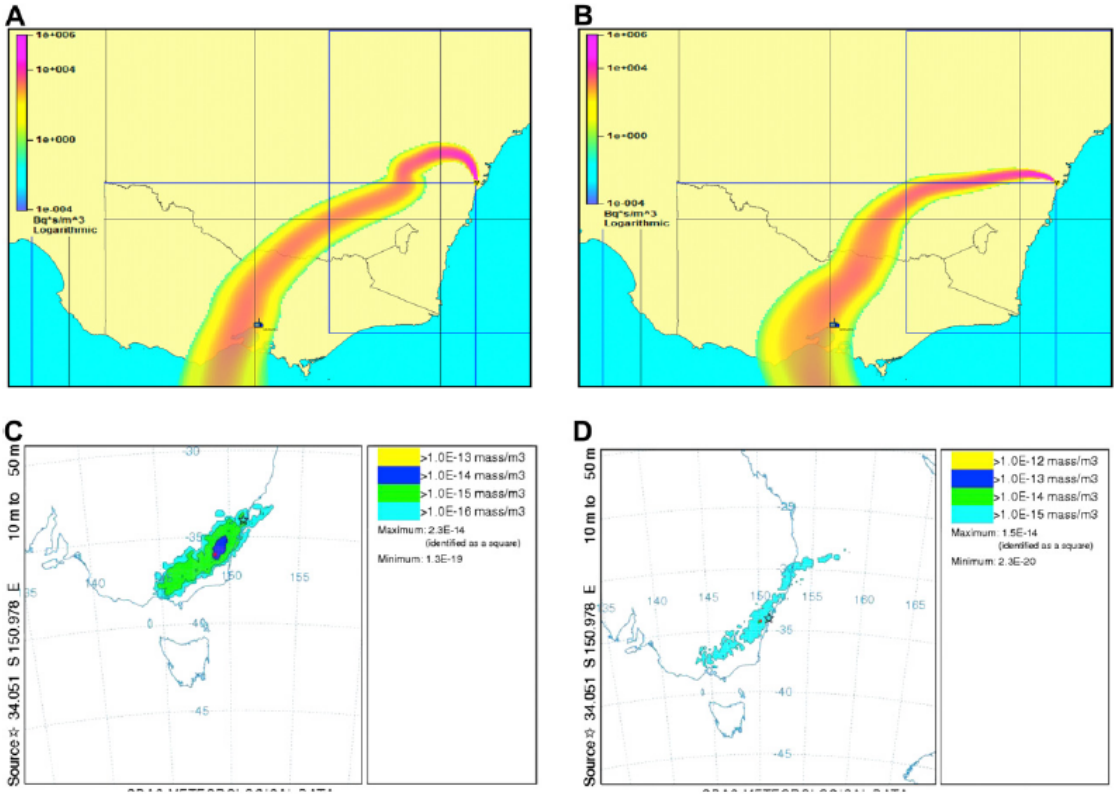

Fig. 3. RIMPUFF integrated concentration for release originating from ANSTO at (A) 24 November 2008 19:30 h and (B) 25 November 2008 06:30 h (icon shows location of Melbourne IMS station). HYSPLIT dilution map for release originating from ANSTO at (C) 24 November 2008 19:00 h and (D) 25 November 2008 7:00 h.

periods in Table 1 are shown in Fig. 4. The MDC of 0.2 mBq m^{-3} was applied to discriminate between predicted activity concentrations of ^{133}Xe that would be detected at the Melbourne IMS station. This value is the average MDC for ^{133}Xe from November 2008 to January 2009. Activity concentrations above the MDC were then used to determine a predicted release activity for each of the detection periods, as shown in Fig. 5. To detect 3 mBq m^{-3} over the first 12 h detection period, the model predicts release magnitudes between 0.5 and 31 TBq. Possible release times are 24 November 7:30–9:30 h, and 24 November 17:30–21:30 h. To detect 6.2 mBq m^{-3} over the second 12 h period would require a release between 0.4 and 34 TBq. Possible release times are 24 November 8:30–10:30 h, 24 November 16:30–20:30 h, and 25 November 6:30–7:30 h. For detection period three, 1.7 mBq m^{-3} was detected

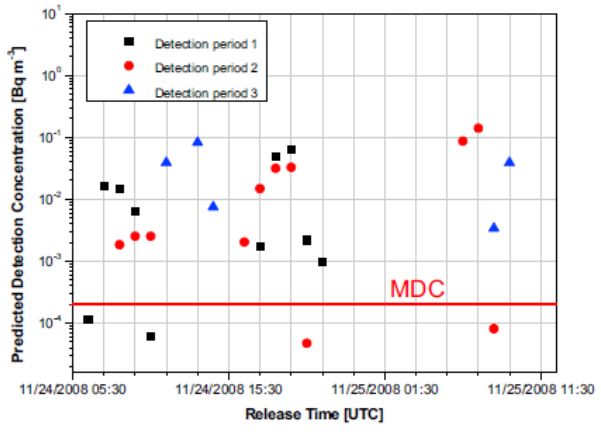

Fig. 4. Predicted detection concentrations in Melbourne for various 10 TBq releases originating from ANSTO. The line at 0.2 mBq m^{-3} indicates the MDC of ^{133}Xe at the Melbourne IMS station.

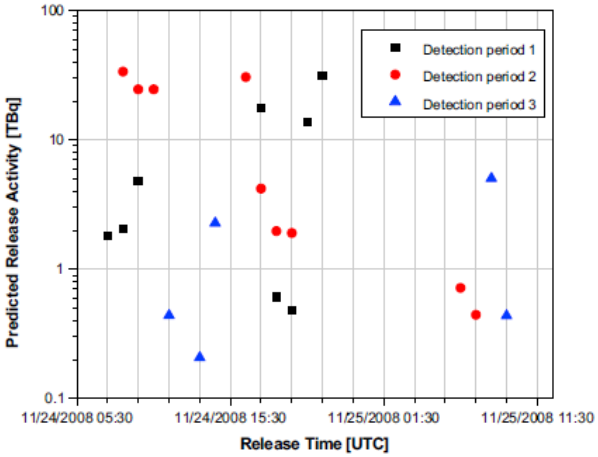

Fig. 5. Predicted release activities from ANSTO based on detected concentrations of ^{133}Xe using the RIMPUFF model.

and the model predicts a likely release between 0.2 and 5 TBq. Possible release times are 24 November 11:30–14:30 h, and 25 November 8:30–9:30 h.

The RIMPUFF forward dispersion analysis predicts ^{133}Xe releases of magnitudes ranging from 0.2 to 34 TBq over a release window from November 24 to 25. This is consistent with the hypothesis of an ANSTO release being responsible for the ^{133}Xe detections by the Melbourne IMS station. Within this time frame the simulations predicted a series of possible single or multiple release times of different magnitudes for each of the three detection periods. Atmospheric dispersion modelling in isolation is not sufficient to uniquely verify the release time. Only comparison to ANSTO stack emission measurements for radioxenon and their isotopic ratios would enable a better understanding of the model predictions.

6. Assessment of ANSTO stack emissions

Following the detection of ^{133}Xe in Melbourne, advice was sought from ANSTO regarding irradiation details and possible source releases from their radioisotope production facility. The stack height of 36 m differed from the 20 m used in the RIMPUFF simulations, but this had no significant effect on the activity concentrations predicted by RIMPUFF. The first hot commissioning trials at the ANSTO radioisotope production facility for ^{99}Mo production took place on 23 November 2008. Stack emission measurements of noble gases are routinely undertaken by ANSTO using a shielded NaI gamma detector in contact with a 100 mL flow-through chamber which samples air exhausted from the ^{99}Mo hotcells immediately after it has passed through High Efficiency Particulate Absorbing (HEPA) and carbon filtration. The gamma spectra are accumulated every 15 min and analysed for the isotopes of interest, then each raw spectrum is added to a weekly sum spectrum for regulatory reporting purposes. Data are available as 15 min, hourly or daily total activity released for ^{133}Xe, ^{135m}Xe, ^{135}Xe, ^{85m}Kr, ^{87}Kr, and ^{88}Kr. Stack emission data for the first hot commissioning trial of ANSTO's new LEU ^{99}Mo plant were supplied for ^{133}Xe and ^{135}Xe and summed for the three days preceding the major detection event in Melbourne in 14 h periods centred around the largest stack releases. These time periods were chosen to obtain sufficient data to determine a ^{135}Xe–^{133}Xe ratio, see Table 2.

The first LEU target was dissolved on 23 November 2008 03:38 h after having been irradiated for 3 d in the local OPAL reactor with the parameters given in Table 3.

To verify how much radioxenon can be expected and what the theoretical isotopic ratios might be, the irradiation of the LEU targets in the ANSTO radioisotope production facility were simulated for one example, the first hot commissioning test run. The simulation tool "Nuclear Analysis 2.0 – Decay and Irradiation Software" (Vilece, 1996) with nuclear data from ENDF/B-VII.0 (ENSDF, 2009) were used.

Fig. 6 shows five different radioxenon isotope activities calculated over time for the first hot commissioning testing. The dashed line indicates the decoupling of the radioxenon isotopes from their parent nuclides due to the target dissolution. The sudden increase in ^{135}Xe after the irradiation stopped (the "xenon poisoning") is noticeable. The figure shows also that around 47 TBq ^{133}Xe was created after the irradiation. The stack emission measurements from 23 to 26 November 2008 for ^{133}Xe ranged from 0.5 to 2.2 TBq, implying that ^{133}Xe emissions were reduced by a factor of around 20. Airborne ^{133}Xe discharges from this facility should not exceed the regulatory notification limit of 280 TBq per annum, equivalent to 5.4 TBq per week (ARPANSA, 2005).

Quarterly dose estimates for all airborne emissions from ANSTO's new ^{99}Mo facility include the noble gases ^{133}Xe, ^{135}Xe, ^{135m}Xe, ^{85m}Kr, ^{87}Kr, ^{88}Kr as well as ^{131}I, ^{132}I and ^{133}I. For the October–December 2008 and January–March 2009 quarters, the estimated maximum effective dose at an on-site receptor location was 0.06 μSv and the off-site estimated maximum was 0.15 μSv in an NNE direction at the 1.6 km ANSTO boundary. These doses are well below the 1 mSv annual limit for public exposure (ARPANSA, 2002). The impact on human health and the environment of radioxenon emissions from ANSTO's production of ^{99}Mo is therefore demonstrably very low, especially considering the average annual dose to a member of the Australian public is 1.5 mSv (Webb et al., 1999).

The ratio of ^{131m}Xe, ^{133m}Xe and ^{135}Xe each to ^{133}Xe over time is shown in Fig. 7. The three horizontal dashed blue lines represent the ratio of ^{135}Xe/^{133}Xe of the three measurements performed in the stack. The upper line crosses the blue simulated line (for ^{135}Xe/^{133}Xe) 1.7 d after cessation of irradiation, the middle blue dashed line after 3 d and the bottom line after 3.5 d. This is consistent, to within a day, with the indicated ANSTO releases described in Table 2. The green horizontal dashed line shows the ^{133m}Xe/^{133}Xe measurement at Melbourne with collection stop 26

Table 2
ANSTO stack emission measurements (without uncertainty) summed over 14 h for major releases of ^{133}Xe and ^{135}Xe.

Date and time	Radioxenon activity inventory (TBq)		
	^{133}Xe	^{135}Xe	^{135}Xe/^{133}Xe
Start: 23 November 2008 19:00 h Stop: 24 November 2008 09:00 h	0.528	0.038	0.072
Start: 24 November 2008 19:00 h Stop: 25 November 2008 09:00 h	2.191	0.015	0.007
Start: 25 November 2008 17:00 h Stop: 26 November 2008 07:00 h	0.746	0.003	0.004

Table 3
Irradiation parameters for the targets dissolved on 23 November 2008.

Neutron Flux [n/cm^{2}s]	9.0×10^{13}
Irradiation time [d]	3.0
Initial Isotope Inventory per target [g]	
^{235}U	20.0
^{238}U	81.3
^{27}Al	280
Number of targets irradiated	8
Cooling time after irradiation [min]	180
Dissolution time [min]	150

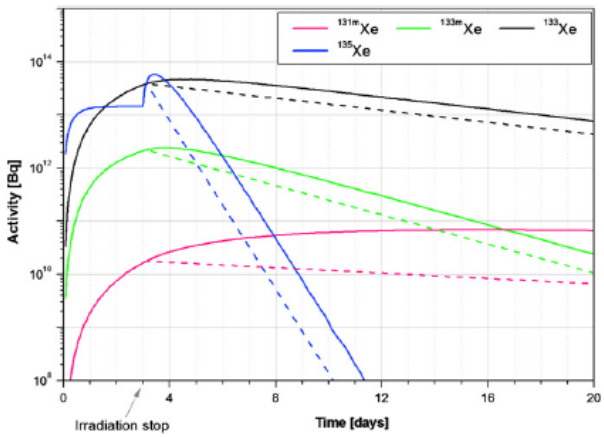

Fig. 6. Results of the simulations for each of the relevant radioxenon isotopes over time given an irradiation time of 3 d where the dashed lines indicate decoupling of the radioxenon from their parent nuclides as a result of the dissolution process.

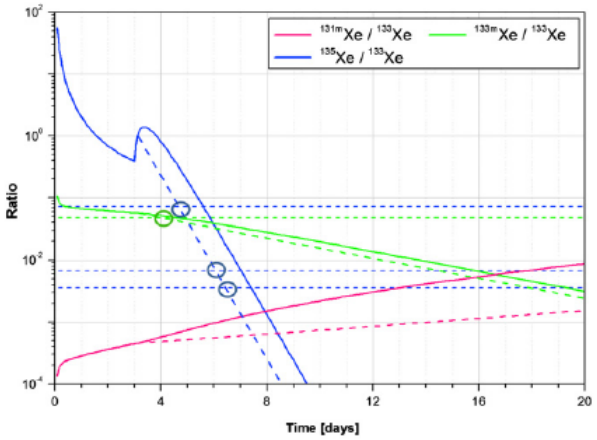

Fig. 7. The ratio of ^{131m}Xe, ^{133m}Xe and ^{135}Xe each to ^{133}Xe over time. Circles indicate intersections between the modelled and measured data (horizontal lines). The three horizontal dashed blue lines represent the ratio of ^{135}Xe/^{133}Xe of the three measurements performed in the stack (Table 2). The green horizontal dashed line shows the ^{133m}Xe/^{133}Xe measurement at the Melbourne IMS station with collection stop 26 November 2008. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

November 2008. This line intersects 1.3 d after cessation of irradiation, corresponding with the known time of the dissolution.

The black line in Fig. 8 shows the simulated ratios of ^{133m}Xe/^{133}Xe and ^{135}Xe/^{133}Xe, and the grey dashed line indicates the decoupled ratios. The red dashed line is the discrimination line proposed by Kalinowski et al. (in press). All measurements right of that line potentially belong to nuclear explosion scenarios or radiopharmaceutical facilities (short irradiation of uranium), whereas everything left is potentially associated with nuclear power plant releases (long irradiation of uranium).

The green dot indicates the Melbourne measurement with its ^{133m}Xe/^{133}Xe uncertainty. The activity concentration of ^{135}Xe was measured below the MDC of 0.48 mBq m^{-3}. Applying this value as an upper limit, the predicted measurement in the 3-isotopic plot must fall in the region shown in light green. The predicted measurement crosses the simulation line between 5.6 and 7 d after irradiation start which corresponds to the earliest plume arrival time at the Melbourne IMS station of 25 November 2008 15:00 h. This is in good agreement with the actual detection, which began during the detection period starting 25 November 2008 19:38 h.

These results show that the release of radioxenon from the ANSTO radioisotope production facility can be simulated correctly, enabling the signal to be differentiated from a possible nuclear test explosion.

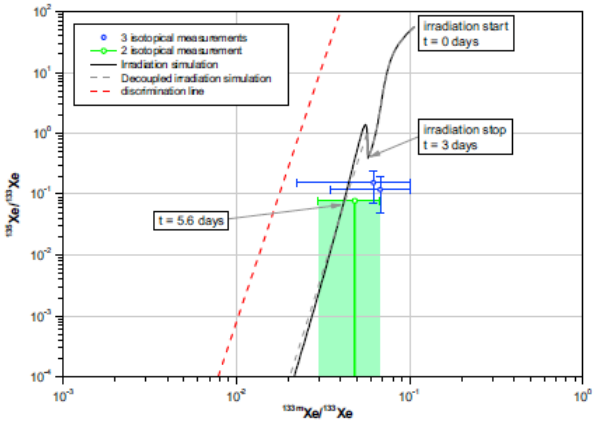

Fig. 8. Ratio of ^{135}Xe/^{133}Xe versus ^{133m}Xe/^{133}Xe. The red dashed line is the nuclear explosion discrimination line, the black line is the simulated ratios of ^{133m}Xe/^{133}Xe and ^{135}Xe/^{133}Xe, and the grey dashed line indicates the decoupled ratios. The green dot indicates the Melbourne IMS station measurement on 26 November with its ^{133m}Xe/^{133}Xe uncertainty. The predicted measurement crosses the simulation line at 5.6 days after irradiation start. The blue points indicate the two measurements on 12 December, in which three radioxenon isotopes were found. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

7. Assessment of other Melbourne detections

From November 2008 to February 2009 the ANSTO radioisotope production facility conducted eight hot commissioning trials for ^{99}Mo production. Fig. 9 shows a time-series plot of radioxenon measurements at the Melbourne IMS station detected over this period, overlaid with the most significant ^{133}Xe ANSTO releases. Four major detections of ^{133}Xe were recorded at the Melbourne IMS station, including the 26 November event already discussed in detail. Application of plume modelling using RIMPUFF within ARGOS enabled each detection event to be linked back to a particular stack emission.

For the maximum detection event on 12 December (Table 1), the modelling output as shown by Fig. 10A, indicates a plume movement in an SW direction, followed by a change to a north direction, before once again moving SSW towards Melbourne. The transport time for this release from ANSTO to Melbourne is estimated at 2 d to correspond to the 10 December ANSTO release. Using the Nuclear Analysis 2.0 – Decay and Irradiation Software the 12 December detections at the Melbourne IMS station were predicted to occur 1–3 d after LEU target dissolution, which occurred on 10 December, this is shown by the blue points in Fig. 8.

For the 21 December maximum, the initial plume movement is in a WNW direction into central New South Wales, eventually moving in an arc south towards Melbourne. The transport time here is estimated to be 2–3 d, corresponding to the 19 December release from ANSTO.

The modelled plume movement for the broad detection event on 28 January is more complex. Initially the plume moves in an SE direction out into the ocean, a change in direction then causes

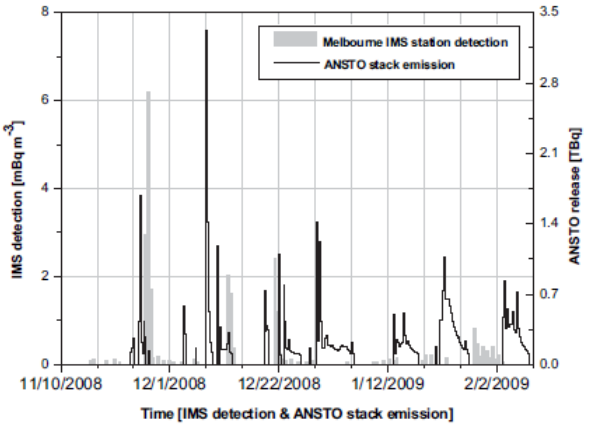

Fig. 9. Time-series plot of ^{133}Xe concentrations (averaged over 12 h) detected at the Melbourne IMS station from November 2008 and February 2009 overlaid with the ANSTO radioisotope production facility ^{133}Xe stack emission release activities in 3 h intervals.

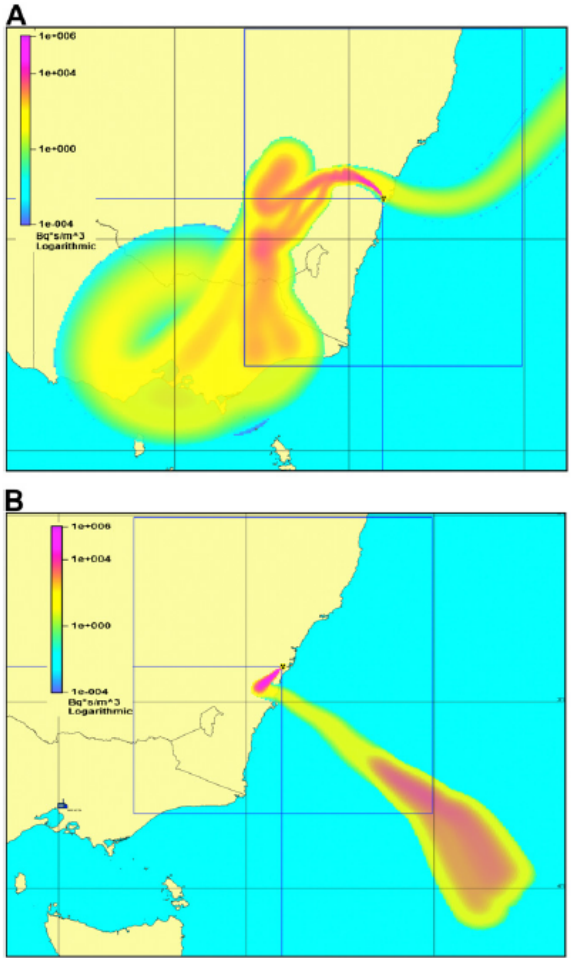

Fig. 10. RIMPUFF transport modelling outputs showing a release at (A) 10 December 2008 5.00 h corresponding to a Melbourne IMS station detection event and (B) for the largest of the stack emission measurements on 8 December 2008.

most of the plume to pass back over Sydney before then travelling in an SW direction towards Melbourne. The transport time is estimated to be 4–6 d, which could correspond to releases from ANSTO starting on 23 January.

The largest stack release from ANSTO, on 8 December, was also modelled (Fig. 10B). In this case the plume movement is in an SE direction out over the ocean and does not reach Melbourne, confirming the scenario of a non-detection of radioxenon at the Melbourne IMS station.

8. Conclusion

The first comprehensive study of radioxenon in Australia successfully applied backward and forward tracking atmospheric dispersion modelling tools to predict source terms and measurements and to connect the November 2008–February 2009 Melbourne radioxenon detections to actual releases from the ANSTO radioisotope production facility. With particular focus on the event at the end of November, the RIMPUFF model within ARGOS predicted single and multiple ^{133}Xe releases ranging from 0.2 to 34 TBq within a 27 h window from 24 November 6:30 h. This was consistent with the ANSTO reported stack emissions of 0.5–2.2 TBq of ^{133}Xe over a similar time period. Application of irradiation modelling coupled with the detection ratios and stack emissions showed that the earliest plume arrival time for LEU targets dissolved on 23 November would be 25 November 15:00 h. The combination of modelling tools applied in this study provides a useful guide to better understanding of their effectiveness in emergency preparedness, source term identification and support of noble gas studies for CTBT treaty verification.

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