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Detection of radioxenon in Darwin, Australia following the Fukushima Dai-ichi nuclear power plant accident

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ABSTRACT

A series of ^{133}Xe detections in April 2011 made at the Comprehensive Nuclear-Test-Ban Treaty Organisation (CTBTO) International Monitoring System noble gas station in Darwin, Australia, were analysed to determine the most likely source location. Forward and backwards atmospheric transport modelling simulations using FLEXPART were conducted. It was shown that the most likely source location was the Fukushima Dai-ichi nuclear power plant accident. Other potential sources in the southern hemisphere were analysed, including the Australian Nuclear Science and Technology Organisation (ANSTO) radiopharmaceutical facility, but it was shown that sources originating from these locations were highly unlikely to be the source of the observed ^{133}Xe Darwin detections.

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1. Introduction

Radioxenon isotopes are short-lived decay products resulting from the nuclear fission process. These isotopes can be released into the atmosphere from radiopharmaceutical facilities, nuclear power and research reactors, and nuclear explosions (Kalinowski and Tuma, 2009; Wotawa et al., 2010). The four key radioxenon isotopes used for detection are ^{131m}Xe ($t_{½}$ = 11.8 d), ^{133m}Xe (2.2 d), ^{133}Xe (5.2 d), ^{135}Xe (9.1 h), with ^{133}Xe and ^{135}Xe isotopes having the highest cumulative fission yields up to 7% (Nichols et al., 2008). Typically ^{133}Xe is the most common isotope to be detected in the environment due to its high yield and long half-life.

Radioxenon is an inert noble gas, therefore it is difficult to contain and is often released into the atmosphere even under controlled conditions. Once in the atmosphere radioxenon is not typically removed through wet or dry deposition processes and can be transported vast distances (Bowyer et al., 2011). These characteristics make radioxenon isotopes ideal tracers to identify nuclear weapons tests. Radioxenon detection stations have been established as part of the International Monitoring System (IMS) of the Comprehensive Nuclear-Test-Ban Treaty Organisation (CTBTO) to detect clandestine nuclear weapons tests. Considerable research has been undertaken to establish criteria for radioxenon isotope activity ratios to allow for discrimination of nuclear weapons tests from civilian sources (Kalinowski et al., 2010). For example, in 2006 elevated levels of radioxenon detected from the IMS station in Yellowknife, Canada, were used to verify an underground nuclear test in North Korea (Saey et al., 2007). Australia maintains seven locations as part of the radionuclide particulate network of the IMS, two of these locations (Melbourne and Darwin) have noble gas detection capabilities (CTBT, 1996).

Radioxenon backgrounds across much of the world are typically in the range of 1–10 mBq m^{-3} (Kalinowski and Tuma, 2009), while regions in close proximity to radiopharmaceutical facilities can be up to a few thousand mBq m^{-3} (Saey, 2009). In Australia, detections of radioxenon up to 10 mBq m^{-3} are routinely made at the Melbourne IMS station. These detections are typically attributed to releases from the Australian Nuclear Science and Technology Organisation (ANSTO) radiopharmaceutical facility (Tinker et al., 2010).

The Fukushima Dai-ichi nuclear power plant (NPP) accident, which occurred in the subsequent days after the earthquake and tsunami off the east coast of Japan on March 11 2011, released large amounts of radionuclides into atmosphere and the ocean. The magnitude of the atmospheric releases of radionuclides attached to particulates have been estimated to be as high as 43% of the Chernobyl accident, with estimates of total radioxenon activity released being more than twice that of Chernobyl (Stohl et al., 2012;

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Schöppner et al., 2012). In the days and weeks following the accident many radionuclide detection systems throughout the northern hemisphere, including the IMS network, experienced highly elevated detections of radioiodine, radiocaesium and radioxenon (Bolsunovsky and Dementyev, 2011; Bowyer et al., 2011; Manolopoulou et al., 2011; Pittauerova et al., 2011).

This study focusses on identifying the probable source of radioxenon detections made in the southern hemisphere from the Darwin IMS station (130.9°E, 12.4°S) in April 2011 using atmospheric transport modelling.

2. Detection

The noble gas detection station located in Darwin as part of the IMS network was installed in November 2006 with a SAUNA II system (Swedish Automatic Unit for Noble gas Acquisition). The system, including detection methods and calculations has been described in detail in Ringbom et al. (2003); a short summary has been provided below. The SAUNA II system samples low volumes of air of less than 20 m^3 every 12 h. Samples are purified by drying and trapping xenon onto charcoal traps. The sample volume is quantified by measuring the stable atmospheric xenon using a thermal conductivity detector and the system is calibrated using internationally traceable xenon isotopes. The concentrations of each of the radioxenon isotopes are measured using a beta-gamma coincidence technique with a measurement time of 11 h 10 min. The average minimum detectable concentration (MDC) of the Darwin station from 2008 to 2010 for ^{133}Xe was 0.2 mBq m^{-3}. The noble gas detection IMS station located in Melbourne is also a SAUNA II system and was installed in October 2008.

A series of ^{133}Xe detections were made by the Darwin IMS noble gas station in April 2011 (Fig. 1). Prior to April 2011, the Darwin IMS noble gas station had not recorded a detection of 1 mBq m^{-3} or greater. Quality assurance was performed for this detection period, to screen out false positive activity concentrations associated with either calibrations or detector problems. There were 17 consecutive detections greater than 1 mBq m^{-3} from 8 April to 16 April 2011. As Table 1 shows, that number of consecutive detections is unique to the noble gas monitoring stations located in Australia. The IMS noble gas station in Melbourne has recorded detections greater than 1 mBq m^{-3}, which can be attributed to releases generated from the ANSTO radiopharmaceutical facility (Tinker et al., 2010). As a means of comparison to the April 2011 series of detections at Darwin, the largest number of consecutive detections made at the Melbourne IMS noble gas station was 5, which occurred in October 2010. It should also be noted that no detections of particulates such as radioiodine or radiocaesium were made during April 2011 at any of the particulate monitoring stations maintained by Australia (Hardman, 2011).

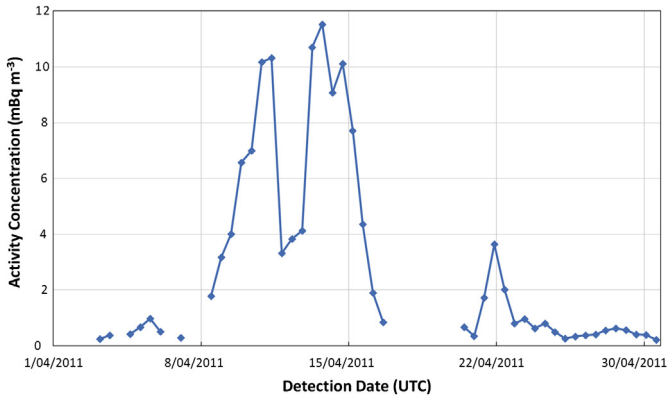

Fig. 1. ^{133}Xe activity concentrations for detections greater than 0.2 mBq m^{-3} in air samples collected from the noble gas IMS monitoring station at Darwin, Australia during April 2011.

Table 1
Consecutive detections for Melbourne and Darwin IMS noble gas stations, pre-Fukushima Dai-ichi NPP accident.

IMS noble gas station	Number of detections >1 mBq m^{-3}	2 or more consecutive detections >1 mBq m^{-3}	4 or more consecutive detections >1 mBq m^{-3}	6 or more consecutive detections >1 mBq m^{-3}
Darwin March 2007–March 2011	0	0	0	0
Melbourne November 2008–March 2011	74	24	3	0

3. Analysis

3.1. Atmospheric modelling

Atmospheric transport models can be used to show the predicted plume passage from a known source, or the potential source regions from a detector point. In this study the Long-range and meso-scale Lagrangian dispersion model FLEXPART (Stohl et al., 2010) has been used. FLEXPART was run in forward mode to track plume movement from Fukushima Dai-ichi NPP site and in backwards mode from the site of the detector. In forward mode the source term of ^{133}Xe chosen was a continuous release of magnitude 10^7 TBq over 5 days starting from 11 March 2011, based on the work of Stohl et al. (2012). The simulations involved the trajectories of 10^7 infinitesimal small air parcels with an output grid of 1° × 1° resolution (latitude × longitude) using meteorological data supplied by the European Center for Medium-Range Weather Forecast.

The output of the FLEXPART backward model is the Source-Receptor Sensitivity (SRS) field, describing the meteorological link for each time interval between each grid cell and the receptor. The total concentration, C (Bq m^{-3}), that is detected during the collection time is given by the sum over all contributing sources. S (Bq) is the emission from a source at the coordinates i and j during the time interval n. M (m^{-3}) is the accordant SRS value (Wotawa et al., 2010).

$$C = \sum M_{ijn} \times S_{ijn}$$

3.2. Source originating from Fukushima Dai-ichi NPP site

The location of the Hadley cells and the Intertropical Convergence Zone (ITCZ) typically acts as a barrier to limit interhemisphere atmospheric transport within the troposphere (UNSCEAR, 2000). In March to April the location of the ITCZ in the Australian-Asia region is very close to the equator (Waliser and Gautier, 1993). However the modelling shows that over a period of around a month, there would be some radioxenon plume movement across the ITCZ into the southern hemisphere. The forward simulation shows that in late March there appeared to be some plume movement into southern Asia and Indonesia (Fig. 2). In the subsequent days and weeks, the modelling shows this air mass had moved into the southern hemisphere through Indonesia into Darwin. There was also some plume movement across the equator through the central Pacific which moved across countries such as Fiji (Fig. 2). According to Avery et al. (2001), March to April is an

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