Journal of Environmental Radioactivity 102 (2011) 681–687

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Elevated radioxenon detected remotely following the Fukushima nuclear accident

T.W. Bowyer [a,*], S.R. Biegalski [b], M. Cooper [a], P.W. Eslinger [a], D. Haas [a], J.C. Hayes [a], H.S. Miley [a], D.J. Strom [a], V. Woods [a]

[a] Pacific Northwest National Laboratory, National Security Division, PO Box 999, Richland, WA, United States
[b] University of Texas, Austin, TX, United States

ARTICLE INFO

Article history:
Received 6 April 2011
Received in revised form
12 April 2011
Accepted 12 April 2011
Available online 21 April 2011

Keywords:
Fukushima
Radioxenon
Xenon
Reactor accident
Xenon-133
Noble gas

ABSTRACT

We report on the first measurements of short-lived gaseous fission products detected outside of Japan following the Fukushima nuclear releases, which occurred after a 9.0 magnitude earthquake and tsunami on March 11, 2011. The measurements were conducted at the Pacific Northwest National Laboratory (PNNL), (46°16′47″N, 119°16′53″W) located more than 7000 km from the emission point in Fukushima Japan (37°25′17″N, 141°1′57″E). First detections of ^{133}Xe were made starting early March 16, only four days following the earthquake. Maximum concentrations of ^{133}Xe were in excess of 40 Bq/m^3, which is more than ×40,000 the average concentration of this isotope is this part of the United States.

1. Introduction

Xenon-133 is a short-lived product of nuclear fission, which is detected in the environment near nuclear facilities such as medical isotope production facilities (Saey et al., 2010), near operating nuclear reactors, and also following nuclear explosions. Radioactive xenon has four isotopes that are commonly detected by many groups and by the International Monitoring System (IMS) of the Comprehensive Nuclear-Test-Ban Treaty (Saey and De Geer, 2005). These isotopes are ^{131m}Xe (11.9 d), ^{133}Xe (5.2 d), ^{133m}Xe (2.2 d), and ^{135}Xe (9.1 h). Xenon-133 is produced near the maximum of the fission yield curve (England and Rider, 1993 and Table 1).

Xenon is an inert noble gas, and therefore is usually one of the first isotopes that can be emitted in processes that produce fission products. As an example, xenon isotopes were detected following the 2006 underground nuclear test in North Korea (Ringbom et al., 2006), and were the highest activity isotopes emitted following the Three Mile Island nuclear accident (Behling and Hildebrand, 1986). Due to its inert nature, xenon is not appreciably scavenged from the atmosphere following a release, and can therefore can travel long distances and be detected with sufficiently sensitive instruments. In addition, the half-lives are nearly optimal for detection because they are short enough that they do not build up in the atmosphere from decay of geological uranium or anthropogenic sources, yet they are long enough to survive wind-borne travel distance from the source at detectable levels.

Sensitive detection of xenon isotopes is commonly performed only after separation of the gas from large volumes of air in order to both remove other interfering gases such as ^{222}Rn and to insert the xenon gas into a favorable counting geometry. Several sources explain the state-of-the-art for this process in some detail (Ringbom et al., 2003). Stable xenon exists in the atmosphere at an abundance of approximately 87 parts-per-billion. Xenon is separated from the major constituents of air, such as N_2, O_2, Ar, CO_2, H_2O, etc., by collecting the gas on adsorbents followed by the use of gas chromatography to further separate the xenon from radon. The xenon gas is then placed into a specialized nuclear detection system for counting radiation emitted as the isotopes undergo radioactive decay. Fig. 1 shows the schematic of a typical radioxenon collection and analysis system.

Radioxenon backgrounds range from well below 1 mBq/m^3 in much of the southern hemisphere (Bowyer et al., 1997) to 1000 mBq/m^3 near medical isotope production facilities. However, most of the world has backgrounds in the range of 1–100 mBq/m^3. In Richland, Washington, where our measurements took place, the background of the xenon isotopes rarely exceeds a few mBq/m^3, because of the lack of local sources.

* Corresponding author. Tel.: +1 509 372 6401.
E-mail address: ted.bowyer@pnl.gov (T.W. Bowyer).

0265-931X/$ – see front matter © 2011 Published by Elsevier Ltd.
doi:10.1016/j.jenvrad.2011.04.009

Table 1
Half-lives and cumulative fission yields of xenon isotopes.

Isotope	Half-Life (days)	Cumulative Fission Yield (%) for ^{235}U
^{131m}Xe	11.9	0.046
^{133}Xe	5.2	6.76
^{133m}Xe	2.2	0.19
^{135}Xe	0.38	6.97

The only known potential source of radioxenon in this part of the world is from the local ~3 GW_{th} Columbi Generating Station (CGS) (12 km NNW of our sampling location), TRIGA reactors at Oregon State University (370 km WSW), Washington State University (165 km WNW) and Reed College (275 km WSW). Winds at the collection time were predominately from the SW, and the TRIGA reactor operators confirmed no detected releases within their facilities, and hence no local source of radioxenon was expected. In the weeks and months preceding the reactor accidents in Japan, PNNL was measuring daily levels of the xenon isotopes, in preparation for a worldwide campaign to measure xenon isotopes at other locations in the world. Xenon-133 is usually the only isotope detected in Richland, though occasionally low levels of ^{135}Xe have been detected, presumably emanated from local sources.

2. The March 2011 accident and the Fukushima nuclear reactors

The Fukushima Daiichi complex of nuclear power reactors consists of six General Electric boiling water reactors (BWRs) units, only 3 of which were reported as operating during a 9.0 magnitude earthquake on March 11, 2011. The nominal power produced by Unit 1 was 1380 MW_{th}, while Units 2 and 3 were both operating at 2381 MW_{th}. Unit 3 has a small admixture of mixed oxide fuel (MOX), such that less than 1% of the fuel was Pu. All other fuel was nominally low enriched uranium (LEU). Immediately after the earthquake, control rods were forced upwards into the core to stop fission reactions, although considerable heat continues to be produced from fission product decay, necessitating continuous circulation of coolant to preserve fuel integrity. Since the reactors were automatically shutdown during or very soon after the earthquake, we have a reliable time in which we believe the reactors were brought to a subcritical state by inserting control rods into the operating reactors. A large inventory of radioxenon and other fission products were still in the fuel after the accident, and then at some time after the tsunami, a thermal runaway occurred and radioactivity was released into the environment. At the time of the writing of this document, few details were available on the amounts of isotopes that were emitted.

Diesel backup equipment present in order to provide cooling water to the reactors was rendered inoperable by the large tsunami that followed a short time later. After batteries were exhausted, cooling was lost, leading to steam generation and a build up of pressure in the pressure vessel. Releases of pressure from the reactor pressure vessels became necessary to keep containment integrity. While the details of the chronology of events are still being determined, one of results of venting of the pressure vessels was the emission of certain fission products. Other releases may have occurred later, for instance in fires that broke out in spent fuel pools of Unit 4, but only gas releases from the pressure vessels could release short-lived radioactive gases.

3. Detections

PNNL has had a 15 + year history in the development of sensitive radioactive xenon (radioxenon) detection systems (Auer et al., 2004; Bowyer et al., 2002; McIntyre et al., 2001; Bowyer et al., 1999),

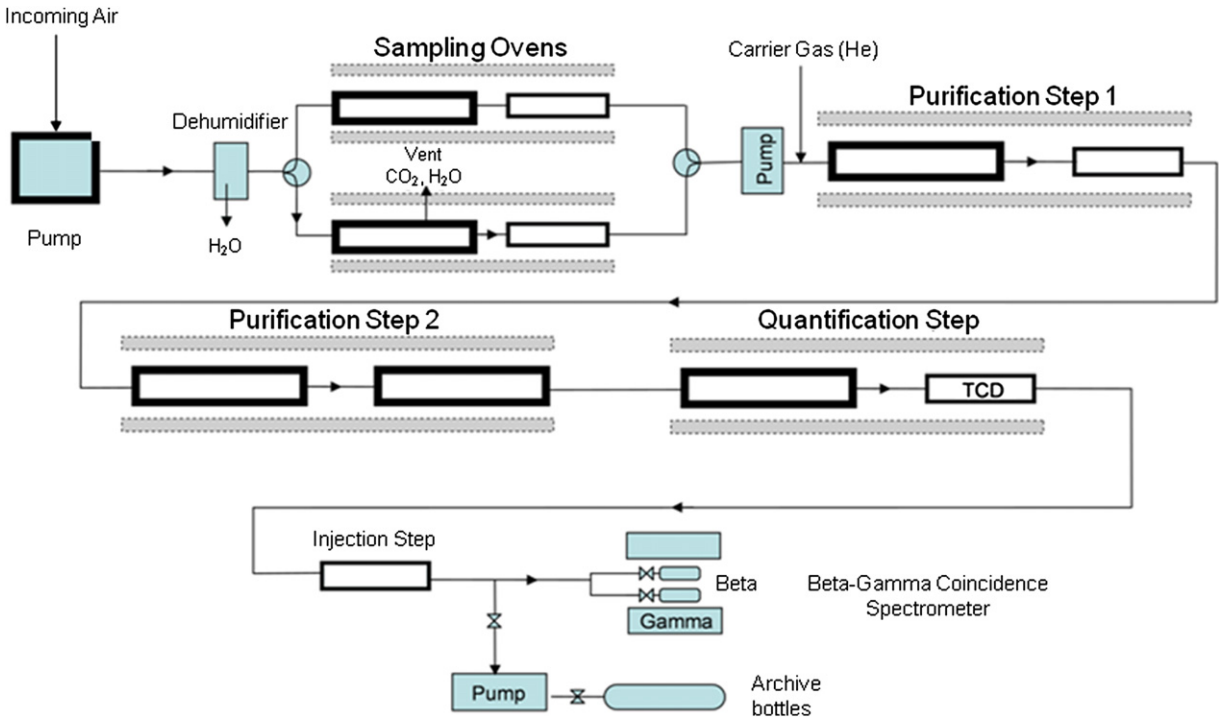

Fig. 1. Schematic illustration of the SAUNA radioxenon measurement system.