The influence of the radioactive background during the temporary suspension of operations of three major radiopharmaceutical production facilities in the Northern Hemisphere and during the start-up of a radiopharmaceutical production facility in the Southern Hemisphere

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Introduction

Atmospheric radioxenon monitoring is a key constituent of the verification of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). Radiopharmaceutical production facilities (RPF) have recently been identified as omitting the major part of the environmental radioxenon measured at globally distributed monitoring sites deployed to strengthen the radiocarbon part of the CTBT verification regime. Efforts on a global xenon measurement inventory revealed that the global total emission from RPF’s is around four times higher than the respective emissions related to maintenance of all nuclear power plants (NPP) worldwide.

Table 1: Overview of the worlds largest RPF’s in 2005 (Bonet et al., 2005).

<table>
<thead>
<tr>
<th>Producer</th>
<th>Country</th>
<th>Production [%]</th>
<th>Days of operations / year</th>
<th>Average release / day [Bq]</th>
<th>Average release / year [Bq]</th>
</tr>
</thead>
<tbody>
<tr>
<td>MEB Nordic</td>
<td>Norden</td>
<td>38</td>
<td>300</td>
<td>1.6 10^9</td>
<td>6.0 10^9</td>
</tr>
<tr>
<td>Mallinkrodt Medical</td>
<td>The Netherlands</td>
<td>26</td>
<td>200</td>
<td>2.5 10^9</td>
<td>7.3 10^9</td>
</tr>
<tr>
<td>IRE</td>
<td>Belgium</td>
<td>16</td>
<td>75</td>
<td>12.0 10^9</td>
<td>10.9 10^9</td>
</tr>
<tr>
<td>NTP</td>
<td>South Africa</td>
<td>16</td>
<td>25</td>
<td>1.3 10^9</td>
<td>4.1 10^9</td>
</tr>
<tr>
<td>Others</td>
<td>Others</td>
<td>4</td>
<td>50</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

In the second half of 2008 the three major RPF’s in the Northern Hemisphere were temporarily shut down due to unrelated reasons. This gave a unique opportunity to see how the noble gas background would behave without these major emitters being operational.

National Research Universal (NRU) in Chalk River, Canada

Nordion at the Chalk River Laboratory facility is the world’s largest producer of 131Mo. Between 24 and 30 August and 4 - 12 December 2008, the reactor was down for scheduled revisions of the installation.

Institut des Radioéléments (IRE) in Fleurs, Belgium

IRE is the third largest 99Mo producer worldwide and the third largest emitter of 133Xe. Between Friday evening 22 August and Monday 25 August 2008, 45 Gbq of 133Xe was released uncontrollably into the atmosphere as a result of the transfer failure of liquid wastes between two tanks. The facility was then shut down on 26 August. On 31 October, IRE received the permission to start up again. Experimental runs were conducted from 13 November on and routine production was resumed from 22 November 2008.

High Flux Reactor (HFR) in Petten, The Netherlands

Mallinkrodt Medical uses targets irradiated from the HFR to produce 99Mo. The reactor was down between 28 July 2008 – 12 February 2009, due to a leakage of the reactor vessel.

Start-up of a radiopharmaceutical production facility in the Southern Hemisphere

In the southern hemisphere the only radiopharmaceutical facility in Australia, the Australian Nuclear Science and Technology Organisation (ANSTO) in Lucas Heights, near Sydney, started production in late November 2008. It uses low enriched uranium targets (19.5% 235U) irradiated at the local 20 MW Open Pool Australian Light-water (OPAL) reactor. Between 23 November 2008 and 4 February 2009, eight hot commissioning runs of the 99Mo production facility were performed. Most emissions occurred on the first day of each of the runs.

2. Environmental observations

The Belgian Telerad network

The Telerad national radiological early warning system, consists of over 200 measurement stations equally distributed over the Belgian territory and focuses on the same time at the Belgian nuclear sites. Around the radiopharmaceutical facility of Fleurus, the network consists of 16 gamma dose rate stations up to about 10 km distance. The effect of the temporary suspension of operations at the site is obvious: during operations, regular peaks in the dose rate, corresponding with radioxenon releases, are observed (Figure 1). These are completely absent during the period of suspension of operations. The maximum dose rate measured at these three stations during operations was 8.95 x 10^-7 Sv/h and during the temporary suspension of operations 1.51 x 10^-7 Sv/h.

Measurements on the Schauinsland IMS station DEX33

In the period before the temporary suspension of operations of Fleurus (24 February 2008 - 25 August 2008), 1261 samples were measured with 133Xe present with an average of 4.5 mBq/m^3. During the temporary suspension of operations of the IRE facility, the average 133Xe went down to 1.1 mBq/m^3 (83 daily samples were measured).

Figure 1: The gamma dose rate (Sv/h) measured at the three measurement stations (IMR/F04, IMR/F05 and IMR/F06) located in the boundary of the site in the dominating wind direction (red contour from the Southeast). The upper graph shows a period of three weeks just before the incident, whereas the lower graph shows the three weeks immediately after the incident.

Measurements in Australia

In the period before the ANSTO RPF start-up, the background of radioxenon at the Melbourne and Darwin IMS stations was below measurable quantities. During the period 7 November 2008 – 7 February 2009, the first radioxenon detections in Melbourne were measured with an average of 0.51 mBq/m^3 133Xe and a maximum of 6.2 mBq/m^3 133Xe. These coincides with the above mentioned pre-production test period production at ANSTO.

Long term continuous radioxenon isotope data from the station on the Schauinsland Mountain were compared with the activities measured before and during the temporary suspension of operations of the main global radiopharmaceutical facilities in the second half of 2008. The average 133Xe activity concentration in the German station decreased with a factor of four. Local measurements in Belgium before and after the temporary suspension of operations in Fleurus, show also a significant decrease of the peak dose rates.

During the downtime of the major two European RPF’s and after the Canadian RPF started up again, the background in Europe showed spikes that in the past probably could not have been seen as they would be covered by the emissions from the Fleurus facility. One spike was influenced by a long distance transport from the Chalk River Laboratory in Eastern Canada, one by a local nuclear power plant and one possibly by a release from a major Russian isotope production facility.

The emissions of a new RPF in Lucas Height are consistent with measurements sampled in Melbourne by analysing the radioxenon isotopic ratios. This is the first major source on the Australian continent.

Both these events, in the Northern and in the Southern Hemisphere clearly prove now the hypothesis and theoretical consideration made in recent papers, that RPF’s are the major contributors to global radioxenon background: when they shut down, the radioxenon background goes down significantly and when they start up in a pristine area, a background is built up.

These releases, however, reduce the sensitivity of the noble gas verification component of the CTBT. It is, therefore, of key importance to search for methods to reduce these radioxenon emissions significantly, as well as in the existing facilities as in the facilities that are being built or are under design.

3. Discussion and outlook

References


Sauer, P. R. J., 3Xe activity concentration, rad/L) at the Melbourne station for the period 7 November 2008 – 28 February 2009. Right: Accumulated 6-hour release activity of 133Xe in the ANSTO chimney. The green vertical line indicates the end of the dissolution of the targets at the ANSTO facility.

Figure 2: 129Xe measurements at the Schauinsland Mountain station for the period 1 June 2008 – 1 January 2009.

Figure 3: combination measurements Schauinsland (30 Sept – 3 Oct) and Schiman (2 Oct am) indicate a release at Chalk River RPF on 24 Sept. as the possible source.