Individual Monitoring Conducted by the Health Protection Agency in the London Polonium-210 Incident

Michael Bailey a*, Alan Birchall a, Louise Bishop b, George Etherington a, Barry Evans c, Graham Fraser b, Roger Gross b, Helen Maguire b, Karen Shaw d, Bernard Wilkins a

a Health Protection Agency, Radiation Protection Division, Chilton Oxfordshire, OX11 0RQ, United Kingdom.
b Health Protection Agency, London Region, 7th Floor, Holborn Gate, 330 High Holborn, London, WC1V 7PP, United Kingdom.
c Health Protection Agency, Centre for Infections, 61 Colindale Avenue, London, NW9 5EQ, United Kingdom.
d Health Protection Agency, South East Epidemiology Unit, 7th Floor, Holborn Gate, 330 High Holborn, London, WC1V 7PP, United Kingdom.

Abstract. Mr. Alexander Litvinenko died on 23 November 2006, having allegedly been poisoned with polonium-210 (210Po) a few weeks earlier [1]. The police investigation identified a number of contaminated locations, including parts of several hotels, restaurants, offices and transport. An extensive programme of individual monitoring of potentially exposed persons was rapidly initiated, based on urine sampling. Methods used for low-level measurement of 210Po in environmental samples were adapted. The Health Protection Agency set a Reporting Level of 30 mBq d⁻¹, results above which indicated likely intake of 210Po from the incident. At each location, risk assessments were undertaken to identify persons with significant risk of contamination with 210Po. These individuals were invited to provide samples, not only to enable a direct assessment to be made of their own exposures, but also to inform decisions on whether others connected with the location should provide samples, or whether they could be reassured. Urine samples from 753 people were processed: about 500 during the first month. Of these, 139 measurements were above the Reporting Level, assessed doses for 36 were in the range ≥1 mSv and <6 mSv, and 17 were ≥6 mSv, with the highest at about 100 mSv. Many of the hotel guests were overseas visitors. An Overseas Advice Team was set up to encourage authorities abroad to adopt similar strategies. Overall, 664 persons from 52 countries and territories were identified. For 176, results of urine measurements were provided to the Overseas Advice Team, of which 13 were above the Reporting Level. Assessed doses for eight of these were <1 mSv, and the other five were in the range ≥1 mSv and <6 mSv.

KEYWORDS: polonium-210, bioassay, emergencies, internal dosimetry.

1. Introduction

Mr. Alexander Litvinenko died on 23 November 2006, having allegedly been poisoned with polonium-210 (210Po) a few weeks earlier [1]. Contamination was found in the two hospitals that had treated him. The police investigation identified more contaminated locations, including parts of several hotels, restaurants, offices and transport. As a result, many staff, guests, customers of these various locations, and visitors to them, were also potentially contaminated. Urine samples were taken from a large number of persons judged at significant risk of contamination with 210Po in order to determine the extent of any contamination.

A description of the overall management of the public health response of the Health Protection Agency (HPA) to the incident is given elsewhere [2]. This paper describes how the urine monitoring programme was set up and conducted, and how the information was used by the HPA to estimate doses to the individuals who provided urine samples. (In this paper ‘dose’ refers to committed effective dose except where qualified.)

* Presenting author, E-mail: mike.bailey@hpa.org.uk
1.1 Biokinetic behaviour of polonium

For general radiation protection purposes the biokinetic behaviour of polonium, after its uptake to blood following absorption from the gastro-intestinal (GI) tract or respiratory tract, is described by the systemic model for polonium recommended by the ICRP (International Commission on Radiological Protection) in Publication 67 [3]. The most notable features are that following uptake to blood, $^{210}\text{Po}$ is widely distributed through soft tissues, but with higher than average concentrations in kidneys, liver, spleen and bone marrow. This is unusual among relatively long-lived alpha-emitters, which generally deposit predominantly in liver and bone. The biological retention half-time following absorption to blood is assumed to be 50 days in all organs, giving an effective half-time of 37 days since the physical half-life of $^{210}\text{Po}$ is 138 days. A recent paper [4] provides an up-to-date review of the behaviour of polonium in the body, and proposes a model that is more detailed and physiologically accurate than the ICRP model. The overall pattern of behaviour is similar, but it includes skin explicitly as a site of retention, and quantifies losses from skin in sweat etc.

1.2 Assessment of Mr. Litvinenko’s intake

An initial assessment was made by HPA staff based on reports in the scientific literature of animal experiments on the acute toxicity of $^{210}\text{Po}$. In particular, Supplement 5 (1964) to the journal Radiation Research, entitled “Metabolism and Biological Effects of an Alpha Particle Emitter, Polonium-210” is a compilation of reports of experiments conducted in the USA. Based on information on the amount of $^{210}\text{Po}$ administered by intravenous injection that would kill rats in various time scales, and assuming that man and rat are of similar sensitivity per kilogram body weight, it was assessed that an initial systemic activity of about 100 MBq would cause death within a few weeks. Using the current ICRP [3] systemic model, uptake to blood of 100 MBq $^{210}\text{Po}$ would give a calculated committed effective dose of 250 Sv. Although not meaningful in relation to the lifetime risk of cancer, it puts the exposure in perspective, as more than 10,000 times the annual limit for workers (20 mSv). The absorbed dose delivered in three weeks to the red bone marrow was calculated to be about 10 Gy, ample to cause severe damage. A comprehensive review of information relating to the acute toxicity of $^{210}\text{Po}$ was subsequently carried out [5], which concluded that uptake to blood of 100 – 300 MBq or more would be fatal to an adult male within 1 month.

Polonium is readily absorbed from the GI tract. The fractional absorption ($f_i$ value) assumed for occupational exposure in ICRP Publication 68 [6], based on consideration of simple inorganic forms of polonium is 0.1. Hence for administration by ingestion, the intake required to give an uptake to blood of 100 MBq $^{210}\text{Po}$ would be of order 1 GBq. The specific activity of $^{210}\text{Po}$ is $1.7 \times 10^{14}$ Bq g$^{-1}$, and so this corresponds to a polonium mass of only 6 micrograms.

1.3 Potential public health hazard

Even 1 GBq $^{210}\text{Po}$ in the body presents no hazard to other people from external irradiation, because $^{210}\text{Po}$ emits only one gamma ray in about 100,000 decays. However, contact with the victim’s body fluids might result in a hazard because of the potential for intakes by inhalation or ingestion. Polonium-210 is widely distributed through body tissues and fluids (section 1.1), and so for a systemic content of 100 MBq, the average concentration in body tissues would be about 1 kBq g$^{-1}$. Since the biological retention half-time in all tissues is 50 days, about 1.5% of the systemic content would be excreted per day, i.e. about 1.5 MBq d$^{-1}$. Of this, one-third is assumed to be excreted in urine (approximately 1500 ml per day) giving about 0.3 kBq ml$^{-1}$. (Excretion in the first few days after intake, especially in faeces, would be considerably greater.) The dose coefficients for $^{210}\text{Po}$ are $2.4 \times 10^{-7}$ Sv Bq$^{-1}$ (ingestion) and $2.2 \times 10^{-6}$ Sv Bq$^{-1}$ (inhalation Type M) [6]. Hence to give a dose equal to the annual limit for workers would require an intake by ingestion of about 80 kBq, or an intake by inhalation of about 9 kBq. These correspond to at least several millilitres of urine or other body fluids. Thus an intake to give such a dose from secondary contamination appeared unlikely, but, given the uncertainties, could not be excluded.
The source material with which Mr. Litvinenko was allegedly poisoned, could however be a much greater potential hazard to people other than the victim. Even 1% of the amount estimated above to have been administered to him might give an effective dose of several Sv. However, the risk from it was much more difficult to assess, because the history of the source material and how it was administered were not known.

2. Initiation of the Individual Monitoring Programme

Environmental (surface contamination) monitoring of places associated with the incident did not start until a few weeks after the presumed date of the poisoning. Therefore, whilst such monitoring provided information on the remaining hazard, from which advice on the need for any remediation measures could be given, it gave only limited information on the original extent of contamination, because of activities such as cleaning which had taken place. Individual monitoring for $^{210}$Po was therefore undertaken, to confirm that doses from secondary contamination from the victim (and perhaps other persons) were low, and to determine any intakes resulting from inadvertent exposure to the source material. It enabled direct assessment to be made of any intakes that had occurred and the resulting individual risks.

A feasibility study showed that individual monitoring based on urine sampling could be used effectively to assess intakes of $^{210}$Po by members of the public and staff at the contaminated locations. Reports in the literature indicated that natural levels of $^{210}$Po in urine from dietary intakes are typically in the range 5 – 15 mBq d$^{-1}$. The collection of information on natural levels of $^{210}$Po in urine has continued. The results of this more extensive survey support the initial conclusions.

2.1 Measurement of Polonium-210 in Urine

A sensitive, but relatively rapid, method was developed for the incident by the Radiation Protection Division (RPD) of the HPA [7], adapted from one in routine use for measurements on environmental samples, e.g. food. In summary, concentrated nitric acid is added to a 1-litre sample of urine, to break down organic matter. The mixture is evaporated slowly (overnight), the residue dissolved in hydrochloric acid, and the $^{210}$Po spontaneously deposited onto a silver metal disc. An alpha spectrometer measures the number of $^{210}$Po alpha particles emitted from the surface of the disc in a suitable period (typically overnight). The minimum detectable activity (MDA) varies between measurements according to the efficiency of the recovery of polonium, which is determined by adding a known amount of a different polonium isotope at the start of the process, and measuring the amount present at the end. During planning it was estimated that an MDA of 20 mBq d$^{-1}$ could be expected routinely. In practice MDAs of 1 – 10 mBq d$^{-1}$ were usually achieved, depending on recovery. During validation of the procedure, measurements were made on urine samples from HPA staff. These confirmed that the procedure was able to measure natural $^{210}$Po levels in both smokers and non-smokers. HPA also carried out validation checks with five laboratories in the UK and eight in other European countries. Some of these laboratories use different methods for the radiochemical isolation of polonium. The results obtained have all been consistent, giving additional confidence in the reliability of the data.

2.2 Choice of Reporting Level

Consideration of natural background levels and the sensitivity of the measurement technique resulted in the choice of a value of 30 mBq d$^{-1}$ for the “Reporting Level” (RL), above which a dose assessment would be made. The dose assessed from a measurement of 30 mBq d$^{-1}$ is only about 1 mSv even for a sample taken three months after intake (section 3.3). Thus it was feasible to confirm that doses were well below the annual dose limit for workers, even for urine samples collected several months after intake.
2.3 Practicalities of Urine Sampling for Large Numbers of People

Since thousands of people had worked in or visited the locations of which parts were contaminated, a sampling strategy was developed to identify those with the highest assessed risk of exposure at each location, and to obtain urine samples from them. This was not only to provide a direct assessment of their own exposure to $^{210}$Po, but also to inform decisions on whether further persons connected with the site should be assessed for providing samples, or alternatively that reassurance could be given to those associated with the site and at a lower risk of exposure.

The immediate requirement to process large numbers of samples, and to report results rapidly, posed some specific challenges including:

- Availability of urine sample bottles in sufficient quantities;
- Instructions on providing 24-hour samples for people whose first language was not English;
- Organisation of transport arrangements at short notice.

Since numbers were likely to exceed the RPD’s analytical capacity of about 40 samples per day, staff contacted other UK laboratories that carry out low-level measurements of $^{210}$Po. Some offered support, which meant rearranging their existing programmes and dedicating laboratory space and equipment to this work. The laboratories were sent check samples to confirm consistency of results, before being sent any real samples. Although about 500 samples were received within the first month, all were analysed in the UK. Important contributions were made by the Veterinary Laboratories Agency (VLA, Weybridge) and the Centre for Environment, Fisheries and Aquaculture Science (CEFAS, Lowestoft) who measured about 200 samples between them in December 2006. As a back-up, several laboratories in Europe were also contacted and sent check samples. No samples were sent abroad as part of the UK measurement programme, but a number of European laboratories analysed samples from their own citizens.

2.4 The Overall Individual Monitoring Process

Public health professional staff from the HPA Local and Regional Services Division (LaRS) London Region were mobilised to undertake public health risk assessments of sites identified as contaminated to a degree that might pose some risk to human health. In conjunction with site management and RPD monitoring teams, individuals connected with the site who were at possible risk of personal contamination with $^{210}$Po were identified. Questionnaires were administered covering occupational, behavioural and temporal factors relevant to potential exposure to environmental $^{210}$Po. Persons assessed as being at significant risk of personal contamination were invited to submit a 24-hour urine sample for measurement of $^{210}$Po activity. Identification of members of the public who were customers at specified sites on certain dates was managed through media calls and triage questionnaires administered by NHS Direct, the UK national healthcare telephone advice service.

Systems were rapidly developed to process hundreds of urine samples each week, including their collection, analysis, dose assessment and reporting. A robust and reliable database was set up to bring together all the information on each sample, to ensure that the correct result was returned to each person, and to provide frequent up-to-date summaries of the programme status and results to the RPD’s Operations Team at Chilton. Integrated reports were regularly prepared with the HPA’s London Region Epidemiology Unit database of assessed persons, for the HPA’s National Emergency Control Centre at Holborn Gate, and thence to central government and the media. These systems were developed and tested in parallel with processing the first samples, the results of which were required urgently to determine whether further measures were needed, and to provide reassurance to persons potentially exposed to $^{210}$Po.

3. Assessments of Intakes and Doses

All calculations described below and carried out by HPA in the monitoring programme used the current ICRP biokinetic and dosimetric models as applied in ICRP Publication 68 [6] i.e., the ICRP Publication 67 polonium systemic model [3], the ICRP Publication 66 Human Respiratory Tract
Model (HRTM) [8], and the ICRP Publication 30 GI tract model [9]. For ingestion, the fractional absorption in the GI tract ($f_i$ value) was assumed to be 10%. For inhalation, absorption Type M was assumed. Calculations below are based on a reference worker (adult male undertaking light work), but similar results would be obtained for other subjects under exposure conditions likely to have been relevant to this incident.

3.1 Urinary Excretion Following Intake of Polonium-210

Figure 1 shows urinary excretion (mBq d$^{-1}$) following an intake of 1 kBq by ingestion or inhalation. To see how the graphs arise, consider someone who ingested 1 kBq 210Po. About 10% of the 210Po is absorbed into blood, and the rest is excreted in faeces within a few days. Of the 100 Bq absorbed, about 0.5% per day is excreted in urine. (As noted above, about 1.5% of the systemic activity is excreted per day: one-third of it in urine). Initially this would result in about 500 mBq d$^{-1}$, but it decreases with time as the amount of 210Po in the body decreases. So, as shown in Figure 1, by the time the first measurements were made (typically about 20 days after presumed intake) urinary excretion is about 300 mBq d$^{-1}$. At 100 days it has fallen to about 100 mBq d$^{-1}$, and by 6 months to about 20 mBq d$^{-1}$.

Figure 1: Urinary excretion following intake of 1 kBq polonium-210 by ingestion, or by inhalation of 5-µm AMAD, 1-µm AMAD or “ambient” aerosols

Inhalation is more complex. Some of the inhaled activity is exhaled again or deposits in the front of the nose, where (based on the HRTM) it is assumed that there is no absorption to blood. Thus, only a fraction of the intake deposits deep enough in the respiratory tract for absorption to blood from it to occur, and this fraction depends on the size of the inhaled particles. Three size distributions were considered and results for them are shown in Figure 1. The AMADs (activity median aerodynamic diameter) of 1 µm and 5 µm are the ICRP default assumptions for exposure of members of the public and workers, respectively. An AMAD of 1 µm is representative of exposures remote from a source, while an AMAD of 5 µm is representative of exposures close to the source of an aerosol produced by dispersion processes (e.g. resuspension of dust). The third size distribution was that of an “ambient” aerosol, which assumes that the 210Po is attached to particles in room air in the same way as radon decay products: most of it is associated with an aerosol of AMAD about 0.2 µm. For the 5-µm aerosol there is high deposition in the upper airways (extra-thoracic and bronchial regions). For progressively smaller particles (1 µm and ambient) there is less deposition in the upper airways and more in the lower respiratory tract (bronchiolar and alveolar-interstitial regions). Hence, if 1 kBq was inhaled, the amount initially absorbed into blood, and hence the excretion in urine, is initially lower than if the
same activity was ingested. However, after inhalation, polonium remaining in the lungs continues to dissolve. For absorption Type M the slow dissolution rate is assumed to be 0.005 d⁻¹, i.e. 0.5% of the activity remaining in the lungs is absorbed into blood each day. Therefore ²¹⁰Po continues to enter the blood, and urinary excretion drops more slowly than after ingestion.

3.2 Intake of Polonium-210 Assessed from a Urine Measurement

As shown in Figure 1, an intake of 1 kBq is predicted to give about 100 mBq d⁻¹ in urine at 100 days, following inhalation or ingestion. It follows that if 100 mBq d⁻¹ in urine is measured at 100 days, then the intake is estimated to be about 1 kBq. As shown in Figure 2, the estimated intake is insensitive to assumptions about the route of intake, especially at times between a few weeks and a few months after intake. A measurement of 100 mBq d⁻¹ made at an earlier time implies a lower intake, and the same measurement made at a later time implies a larger intake.

**Figure 2:** Intake by ingestion, or by inhalation of 5-µm AMAD, 1-µm AMAD or “ambient” aerosols, estimated from a measurement of 100 mBq d⁻¹ polonium-210 in urine

3.3 Dose Assessed from a Urine Measurement

The dose calculated for intake of 1 kBq by ingestion is 0.24 mSv. The doses calculated for intakes of 1 kBq by inhalation are 2.2, 3.0 and 3.3 mSv for 5-µm AMAD, 1-µm AMAD and ambient aerosols, respectively. Doses from inhalation are much higher than for ingestion because of the additional dose to the lungs, which are sensitive to radiation-induced cancer, and so assigned a high tissue weighting factor. The dose assessed from a measurement of 100 mBq d⁻¹ at any time is obtained by multiplying the intake (Figure 2) by the dose per kBq: results are shown in Figure 3. Thus, assuming 100% intake by inhalation, a measurement of 100 mBq d⁻¹ at about 100 days gives an assessed dose in the range 1 – 5 mSv, depending on the aerosol size. Assuming 100% ingestion the dose is much lower. Figure 3 also shows results assuming that intake was a mixture of ingestion and inhalation, because a mixture was assumed in most special assessments (see below). For a measurement different from 100 mBq d⁻¹, the result is simply scaled in proportion. As shown in Figure 3, the assessed dose depends on assumptions made about the route of intake, and for inhalation, on the aerosol size, but is not very sensitive to the assumed time of intake.
Figure 3: Dose assessed from a measurement of 100 mBq d\(^{-1}\) polonium-210 in urine, for intake by ingestion, by inhalation of 5-µm AMAD, 1-µm AMAD or “ambient” aerosols, or from a mixture (50% inhalation of 1 µm aerosol and 50% ingestion)

3.4 Dose Assessment Procedure for Response to the Polonium-210 Incident

Because of the large number of samples expected, and the need for rapid reporting of results, a system was developed by which rapid assessments would be made for those individuals whose urine measurements indicated that their intakes and doses were negligible, while thorough assessments would be made for those individuals likely to have received greater intakes and doses. Thus, if the measurement was less than the Reporting Level (RL) of 30 mBq d\(^{-1}\), no dose assessment was carried out, and the result was reported as “Below Reporting Level” (Category 1). If the measurement was above the RL, a standard assessment was carried out on the cautious assumption of 100% inhalation. If this gave a dose less than 1 mSv, it was reported simply as “<1 mSv” (Category 2). If, however, the standard assessment gave a dose greater than 1 mSv, a special assessment was carried out (Category 3). Information obtained during the initial risk assessment process was used to inform judgements about the potential for exposure by both routes of intake. In most cases a mixture of inhalation and ingestion was assumed. In reporting results to those without radiation protection expertise, emphasis was placed on giving a clear and simple message. Doses assessed to be <6 mSv (Category 3a, as well as Categories 1 and 2) were described as being “of no concern”, and doses assessed to be ≥ 6 mSv (Category 3b) were described as being “of some concern” [7].

4. Results of the UK Monitoring Programme

Samples from a total of 753 people were processed: 500 in the first month or so (Table 1). Out of these, 139 measurements (18%) were above the RL, and therefore indicated likely intakes of \(^{210}\)Po associated with the incident. The proportion was lower (13%) in healthcare workers (mainly staff at the two hospitals where he was treated) than in the others who provided samples (19%). The latter were potentially exposed to \(^{210}\)Po in a variety of locations (Table 2). Of results above the RL, assessed doses for 36 were in the range ≥ 1 mSv and <6 mSv (Category 3a), and 17 were ≥ 6 mSv (Category 3b), with the highest at about 100 mSv.
Table 1: Results of measurements of 24-hour $^{210}$Po urinary excretion

<table>
<thead>
<tr>
<th></th>
<th>Samples assessed</th>
<th>Below Reporting Level</th>
<th>$30 \text{ mBq d}^{-1}$ to $&lt;1 \text{ mSv}$</th>
<th>$\geq 1 \text{ mSv and } &lt;6 \text{ mSv}$</th>
<th>$\geq 6 \text{ mSv}$ Total presumed contact with $^{210}\text{Po}$</th>
<th>Percentage presumed contact with $^{210}\text{Po}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Healthcare</td>
<td>78</td>
<td>68</td>
<td>9</td>
<td>1</td>
<td>0</td>
<td>10</td>
</tr>
<tr>
<td>Non-Healthcare</td>
<td>674(^a)</td>
<td>545</td>
<td>77</td>
<td>35</td>
<td>17</td>
<td>129</td>
</tr>
<tr>
<td>Total</td>
<td>752</td>
<td>613</td>
<td>86</td>
<td>36</td>
<td>17</td>
<td>139</td>
</tr>
</tbody>
</table>

\(^a\) One further result in this group was not categorised, and is not included in the table. Polonium-210 was not detected, but because of the small sample volume, the result could only be reported as $<100 \text{ mBq d}^{-1}$.

Table 2: Results of measurements of 24-hour $^{210}$Po urinary excretion above Reporting Level

<table>
<thead>
<tr>
<th>Group</th>
<th>No of results above Reporting Level (presumed contact with $^{210}\text{Po}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Healthcare</td>
<td>10</td>
</tr>
<tr>
<td>Family, friends, visitors</td>
<td>10</td>
</tr>
<tr>
<td>Offices (staff and visitors)</td>
<td>8</td>
</tr>
<tr>
<td>Restaurant staff</td>
<td>7</td>
</tr>
<tr>
<td>Hotel staff</td>
<td>56</td>
</tr>
<tr>
<td>Hotel guests</td>
<td>10</td>
</tr>
<tr>
<td>Bar customers</td>
<td>38</td>
</tr>
<tr>
<td>Total</td>
<td>139</td>
</tr>
</tbody>
</table>

5. Monitoring of Persons Overseas and International Liaison

It was soon apparent that many of the people potentially exposed at locations found to be contaminated were visitors from overseas. An Overseas Advice Team (OAT) was established with up to seven staff from the HPA’s Centre for Infections and LaRS [10]. The OAT identified visitors from overseas at contaminated locations: 460 from 52 different countries and territories, making up a significant proportion of the total identified potentially exposed population. A further 204 persons self-identifying themselves as present at contaminated venues were also reviewed. The OAT contacted and liaised with public health authorities in these persons’ home countries, and encouraged sampling of those with highest potential exposure, as for UK residents. Documentation describing the principles of the public health risk assessment and urine sampling programme in the UK were provided. RPD staff worked closely with the OAT, providing technical support and where appropriate using existing contacts with overseas professional colleagues. RPD staff also provided advice to other countries on sampling and dose assessments where requested.

The OAT actively sought the results of measurements carried out overseas to compile a database complementary to that of the measurements made in the UK, since all results provide information about conditions at the various contaminated locations that assists in assessing the exposures of everyone who was there. Of the 168 results obtained, 13 were greater than the RL, eight giving assessed doses less than 1 mSv, and five in the range 1 to 6 mSv.

4. Conclusions

The rapid development of effective systems for identifying those individuals at the highest risk of significant intakes from the thousands potentially exposed, collecting urine samples from them, measuring the $^{210}$Po present, assessing their doses and communicating the results, required enormous effort in a short space of time. Many Health Protection Agency staff, mainly in Radiation Protection
Division and Local and Regional Services but also in other Divisions were involved. The combined resources of Health Protection Agency enabled an effective response to be made to an extraordinary event.

Acknowledgements

Important contributions were made by the Centre for Environment, Fisheries and Aquaculture Science (CEFAS, Lowestoft) and the Veterinary Laboratories Agency (VLA, Weybridge) who between them measured about 200 urine samples during December 2006. Many members of staff in the Health Protection Agency, particularly in the Radiation Protection Division and Local and Regional Services Division, contributed to the work reported here. It is not possible to recognise them all individually, but their contributions are gratefully acknowledged.

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