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

M R Bailey[a], A Birchall[a], G Etherington[a], G Fraser[b], B T Wilkins[a], Y Bessa[a], L Bishop[b], J Brown[a], M-D Dorrian[a], L W Ewers[a], T P Fell[a], G J Ham[a], D J Hammond[a], S Forrester[b], A Hodgson[a], C Howarth[a], Z-F Ibrahimi[a], H Maguire[b], J Marsh[a], A W Phipps[a], M Puncher[a], R Ruggles[c], A Shutt[a], J R H Smith[a], D Turbitt[b], M J Youngman[a] and D Wilding[a]

[a]RADIATION PROTECTION DIVISION
[b]LOCAL AND REGIONAL SERVICES DIVISION
[c]CHEMICAL HAZARDS AND POISONS DIVISION

ABSTRACT

The alleged poisoning of Mr Alexander Litvinenko with polonium-210 was an extraordinary event that presented some unique public health challenges. Environmental polonium-210 contamination was found at a number of locations in London, including parts of two hospitals, several hotels, restaurants, and office buildings. An extensive programme of individual monitoring of potentially exposed persons was rapidly initiated, based on urine sampling. At each location, risk assessments were undertaken to identify persons with significant risk of contamination with polonium-210. These individuals were invited to provide samples, not only to enable a direct assessment to be made of their own exposure, but also to inform decisions on whether others connected with the site should also provide samples or could be reassured. Urine samples from 753 people were processed: about 500 during the first month, another 250 up to the end of May 2007, and a further three up to August 2007. Of these, 139 measurements were above the Reporting Level set by the Health Protection Agency for this incident of 30 mBq d^{-1}, showing the likely presence of polonium-210 from the incident. Committed effective doses were assessed for measurements above the Reporting Level. Most were less than 1 mSv, with thirty-six in the range ≥ 1 mSv and <6 mSv, and seventeen ≥ 6 mSv, with the highest about 100 mSv.

Centre for Radiation, Chemical and Environmental Hazards
Radiation Protection Division
Chilton, Didcot, Oxfordshire OX11 0RQ

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The following amendment has been made to this document since its first publication (June 2010):

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Section 1.2 (Page 3): Original text “(1 microcurie, here written µc, but more usually abbreviated to µCi, is equal to 37 MBq)” corrected to “…equal to 37 kBq)”.

EXECUTIVE SUMMARY

Mr Alexander Litvinenko died on 23rd November 2006, having been allegedly poisoned with the radionuclide polonium-210 (^{210}Po) a few weeks earlier. Contamination was found in the two hospitals that had treated him. Over the following days the scope of the incident enlarged and diversified as the police investigation identified more contaminated locations in London, including parts of several hotels, restaurants, and office buildings.

According to the current model for the systemic behaviour of polonium (i.e., its behaviour after uptake to blood) recommended by the International Commission on Radiological Protection (ICRP), polonium is widely distributed through soft tissues, but with higher than average concentrations in kidneys, liver, spleen and bone marrow. The assumed biological retention half-time in all tissues is 50 days, with one third of the excretion going to urine. Polonium is readily absorbed from the gut: fractional absorption is taken to be 10% for simple inorganic forms. A recent (2001) paper by Leggett and Eckerman 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.

An initial assessment based on reports in the literature of the toxicity of ^{210}Po in animals, and consideration of tissue doses, indicated that systemic uptake (absorption to blood) of the order of 0.1 GBq ^{210}Po could cause death in a few weeks. This suggested that the amount administered would have been of the order of 1 GBq, equivalent to 6 μg polonium. Since intakes of order 10 kBq are needed to give committed effective doses exceeding the annual limit for workers (20 mSv) it was considered unlikely that exposures giving rise to such doses would have arisen through secondary contamination from the victim. However, the source material with which Mr Litvinenko was allegedly poisoned posed a much greater potential hazard.

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}. Methods used for low-level measurement of ^{210}Po in environmental samples were adapted and tested for analysis of urine samples. It was shown that levels of 20 mBq d^{-1} could readily be measured on a routine basis. This would enable doses of the order of 1 mSv to be assessed, even if samples were obtained several months after intake. On that basis a Reporting Level of 30 mBq d^{-1} was set, measurements above which were considered likely to include some contamination from the incident.

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.

Public health professional staff from the Health Protection Agency 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 Radiation Protection Division (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 (NHSD), the UK 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 Agency's London Region Epidemiology Unit database of assessed persons, for the Agency'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.

With support from other UK laboratories, samples from about 500 people were processed in the first month, another 250 were processed up to the end of May 2007, and a further three up to August 2007. Of these, 613 results were below the Reporting Level of 30 mBq d^{-1}, and most of these were below 20 mBq d^{-1}, consistent with the expected range of natural background for people in the UK. One hundred and thirty-nine results (18.5%) were above the Reporting Level, showing the likely presence of some ^{210}Po from the incident. Of these, 92, 41 and 6 were in the ranges 30–100, 100–1000 and >1000 mBq d^{-1}, respectively.

In addition to the public health programme, 24 measurements were made on urine samples from 17 members of Health Protection Agency staff. Three were control samples and the rest followed involvement in the environmental monitoring programme. Of these, 23 results were below the Reporting Level.

For results above the Reporting Level, biokinetic models were used to calculate (i) the intake, the amount of ^{210}Po that originally entered the body that would give such an excretion rate, and (ii) the radiation (committed effective) dose resulting from that intake. However, the assessed doses depend on assumptions about the exposure – in particular inhalation gives a higher dose than ingestion.

Since the models used assume that about 10% of the intake is absorbed into blood whether by inhalation or ingestion, the intake assessed from a urine measurement is not

very dependent on assumptions about the route of intake. An intake of 1 kBq ^{210}Po is predicted to give about 100 mBq d^{-1} in urine at 100 days after intake by either route. The intake assessed from a measurement of 100 mBq d^{-1} increases from about 0.3 kBq for a measurement at 20 days after intake to about 5 kBq for a measurement made 6 months after intake. The committed effective dose calculated from intake of 1 kBq, is, however, about 10 times higher for an intake by inhalation than for an intake by ingestion. The dose assessed from a urine sample is therefore much higher if inhalation rather than ingestion is assumed. For samples taken in the first few weeks after intake it also depends on the assumed size distribution of the inhaled particles.

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 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 Reporting Level, 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. 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".

Out of 139 measurements that were above the Reporting Level, and therefore indicating likely intakes of ^{210}Po associated with the incident, 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. Results above the Reporting Level were measured in people exposed in many locations.

For specific groups of people (designated by location and occupation) the proportion of results above the Reporting Level was relatively high (>35%) in four: Mr Litvinenko's family and friends; office staff; guests at one hotel; and hotel bar staff. In other groups it was broadly similar (about 5 – 25%). The proportion of Category 3 results was relatively high (>15%) in the same four groups. In most others it was broadly similar (about 3 – 10%), but it was relatively low in health care workers (1%). Many of the Category 3 results (26 out of 53) were associated with potential exposures in a hotel bar (either staff or customers) including 11 out of the 17 in Category 3b.

Many people potentially exposed at contaminated locations were visitors from overseas. The Health Protection Agency established an Overseas Advice Team (OAT) to identify and follow up such individuals, and to assist authorities to conduct assessments based on criteria developed for UK residents. Overall, 664 persons from 52 countries and territories were identified. For 176, results of urine measurements were provided to the

OAT. Individuals were placed into three categories of potential exposure risk ('higher', 'lower' and 'unknown') based on available information. Results for 19% of those at 'higher' risk were above the Reporting Level, a similar proportion to that in UK residents tested. For those at 'unknown' or 'lower' risk the fraction was 3%.

Evidence supporting the assumption that results above the Reporting Level of 30 mBq d^{-1} indicate likely contact with ^{210}Po in this incident comes from the low proportion of results above the Reporting Level observed in a group of restaurant customers (0%), Health Protection Agency staff (4%), and overseas visitors at low or unknown risk (3%).

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 the Health Protection Agency enabled an effective response to be made to an extraordinary event.

CONTENTS

1 INTRODUCTION

Mr Alexander Litvinenko died on 23 November 2006, having allegedly been poisoned with polonium-210 (^{210}Po) a few weeks earlier (Hansard, 2007). Contamination was found in the two hospitals that had treated him. The police investigation identified more contaminated locations, including parts of several hotels, restaurants, and office buildings. As a result, many staff, guests, customers of, and visitors to, these various sites were also potentially contaminated. Twenty-four-hour urine samples were taken from a large number of persons judged at significant risk of contamination with ^{210}Po (over 750) in order to determine the extent of any contamination.

This report describes how the urine monitoring programme was set up and conducted, and how the information was used by the Health Protection Agency to estimate the committed effective doses to the individuals who provided urine samples. (In the rest of this document 'dose' refers to committed effective dose except where qualified.) Summaries of this work have been presented previously, most aimed primarily at the general public and public health professionals (HPA 2007a, 2007b, 2007c, Bailey et al 2008). This report provides more technical detail mainly to inform radiation protection professionals. However, additional explanation is given in places to make it more accessible to those without training in radiation protection generally or internal dosimetry specifically. Summaries of the Health Protection Agency's overall public health response to the incident are given elsewhere (HPA 2007a, Croft et al 2008, Maguire et al in press, Shaw et al in press).

1.1 Initial assessment of the situation

Experts from the Health Protection Agency's Radiation Protection Division (RPD) were initially tasked to:

- consider whether it was plausible that Mr Litvinenko's symptoms could be caused by an intake of ^{210}Po, and if so:
    - estimate how large an intake he might have received;
    - make an initial assessment of potential risks to those who were in contact with him from the presumed time at which he was poisoned, i.e. the public health consequences;
- collect information on natural levels of ^{210}Po in human body tissues and excreta.

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 (ICRP, 1993) (Figure 1). It is based on the simple empirical model adopted earlier in ICRP Publication 30 (ICRP, 1979), but with the explicit inclusion of excretion pathways to urinary bladder and the GI tract. The most notable features are that following uptake to blood, ^{210}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. Following ingestion, the highest doses to tissues with specific tissue weighting factors are to kidney, liver, and bone marrow. Polonium is also readily absorbed from the GI tract. The fractional absorption (f_1 value) assumed for occupational exposure in ICRP Publication 68 (ICRP, 1994a), based on consideration of simple inorganic forms of polonium is 0.1. The f_1 value assumed for environmental exposure of adult members of the public in ICRP Publication 67 (ICRP, 1993) based on consideration of polonium biologically incorporated in food is 0.5. The biological retention half-time following absorption to blood is assumed to be 50 days in all organs. Since the physical (radioactive decay) half-life of ^{210}Po is 138 days, the overall (effective) retention half-time in the body after systemic uptake is $(1/50 + 1/138)^{-1}$ = 37 days.

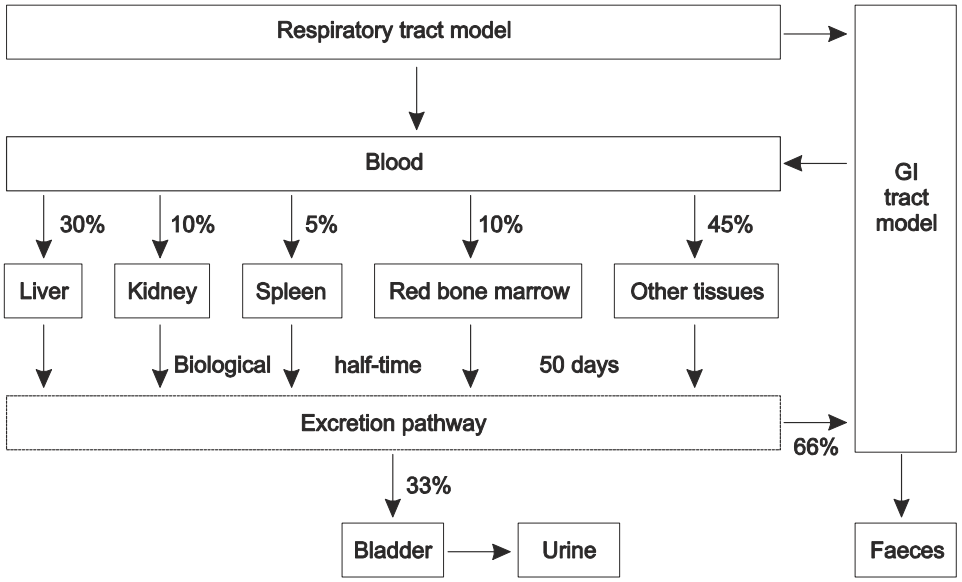

Figure 1 ICRP Publication 67 (1993) Polonium Systemic Model

A more recent, physiologically-based systemic model for polonium (Figure 2) was published by Leggett and Eckerman (2001), and is currently under consideration by the ICRP Task Group on Internal Dosimetry in its review of systemic models, in the development of a new document on Occupational Intakes of Radionuclides. The overall pattern of distribution, retention and excretion is broadly similar to that represented by the Publication 67 model. The main differences from the ICRP Publication 67 model of importance here are:

- explicit inclusion of skin as a site of deposition, and loss from skin to sweat, etc;
- somewhat lower urinary excretion following systemic uptake through injection or ingestion;

- greater urinary excretion following uptake from the respiratory tract (through the compartment labelled “Plasma 2”), than from the GI tract or direct injection into blood.

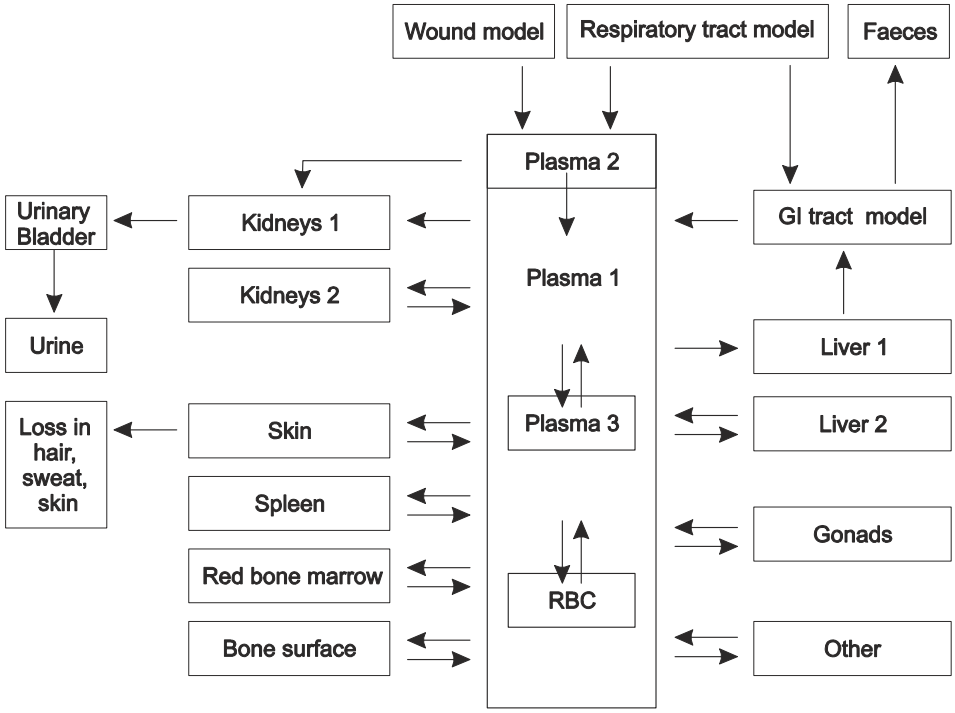

Figure 2 Leggett & Eckerman (2001) Polonium Systemic Model. Reproduced from Science of the Total Environment, Volume 275 pp109-125. RW Leggett and KF Eckerman, A systemic biokinetic model for polonium. Copyright 2001, with permission from Elsevier.

1.2 Acute toxicity of polonium-210

There are reports of animal experiments on the acute toxicity of ^{210}Po in the scientific literature. In particular, Supplement 5 to the journal Radiation Research, which is entitled “Metabolism and Biological Effects of an Alpha Particle Emitter, Polonium-210” is a compilation of experiments conducted at the University of Rochester. Of particular relevance within it are papers by Cassarett (1964) on the effects of single intravenous injections of ^{210}Po, and by Della Rosa and Stannard (1964) on the acute toxicity of ^{210}Po as a function of route of entry. Both refer to earlier work reported by Fink (1950), according to which, rats administered “dosages of 120 or 170 µc/kg died in about a week and showed aplastic marrow” (1 microcurie, here written µc, but more usually abbreviated to µCi, is equal to 37 MBq). The dosage by intravenous injection into rats for 50% lethality in 20 days was reported to be 43 µc/kg and for 50% lethality in 40 days was reported to be 27 µc/kg. On the preliminary assumption that man and rat are of similar sensitivity, an administered activity of 40 µc/kg would be likely to cause death in a few weeks. Scaled up to a 70-kg man, this gives an initial systemic activity of about 100 MBq.

Using the current (ICRP Publication 67) systemic model, uptake to blood of 100 MBq ^{210}Po would give a calculated committed "effective dose" of 250 Sv. Although this is not meaningful in relation to the normal use of effective dose as an approximate measure of the lifetime risk of late effects, notably cancer, it puts the exposure in perspective as giving more than 10,000 times the annual dose limit for workers (20 mSv). For the red bone marrow, the following absorbed doses can be calculated, absorbed doses being more relevant to acute effects than equivalent or effective dose:

- committed absorbed dose = 26 Gy (formally dose absorbed in 50 years after intake);
- absorbed dose in 3 weeks = 9 Gy;
- initial dose rate = 0.5 Gy per day.

This does not consider the relative biological effectiveness (RBE) for alpha particle irradiation with respect to bone marrow damage, which is likely to be greater than unity, but less than the radiation weighting factor of 20 adopted by ICRP for the calculation of equivalent dose. A comprehensive review of information relating to the acute toxicity of ^{210}Po has since been carried out (Harrison et al, 2007), considering other reported studies of ^{210}Po toxicity, notably in species other than rats, and information on acute radiation effects on those human tissues that are relevant to this case. Nevertheless, the brief assessment above demonstrates that poisoning with ^{210}Po is a plausible hypothesis, and gives an order of magnitude estimate of the amount that might have been involved.

For simple inorganic forms of ^{210}Po, the fractional absorption in the GI tract (f_1 value) is taken to be 0.1 (ICRP, 1994a). Hence for administration by ingestion, the intake required would be of order 1 GBq. The specific activity* of ^{210}Po is 1.7 x10^{14} Bq g^{-1}, and so this corresponds to a polonium mass of only 6 micrograms.

1.3 Potential public health hazard

Even with up to 1 GBq in the victim's body, there would be no hazard to other people from external irradiation, because ^{210}Po is almost a pure alpha-emitter, with only a very low yield gamma-emission (0.00121%) at 803.10 keV (Brookhaven National Laboratory, 2007). However, contact with body fluids, directly or from the contaminated environment, might result in a hazard because of the potential for intakes by inhalation or ingestion.

* The number of atoms in 1 gram-molecular weight of any compound is given by Avogadro's number: $N_A = 6.023 \times 10^{23}$. Thus for pure ^{210}Po, 210 grams = 6.023×10^{23} atoms, and 1 gram = 2.9×10^{21} atoms. The decay rate (the probability that an atom will decay during 1 second), $\lambda = \ln(2)/t_{½}$, where $t_{½}$ is the radioactive half-life = 138 days = 138 x 24 x 3600 s = 1.19×10^7 s.
Hence $\lambda = \ln(2)/t_{½} = 5.8 \times 10^{-8}$ s^{-1} and the number of decays per second from 1 gram is $(2.9 \times 10^{21}) \times (5.8 \times 10^{-8}) = 1.7 \times 10^{14}$.

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 per gram.

According to the ICRP Publication 67 systemic model, the biological retention half-time in all tissues is 50 days, so about 1.5% of the systemic content (100 MBq) is excreted per day, i.e. about 1.5 MBq per day. 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 Leggett and Eckerman (2001) model identifies sweat as another excretion pathway (Figure 2), although it is not separated from other losses from skin. The model predicts that these combined losses from skin are in the range 10 – 50% of the excretion in urine in the period 10 – 140 days after intake by ingestion.

A simple, scoping assessment of the doses that could result from intake of ^{210}Po in body fluids was made by considering the dose coefficients (i.e., committed effective dose per unit intake) for inorganic compounds from ICRP Publication 68, which are 2.4×10^{-7} Sv Bq^{-1} (ingestion) and 2.2×10^{-6} Sv Bq^{-1} (inhalation Type M). Hence to give a committed effective dose equal to the dose limit of 20 mSv for workers would require an intake by ingestion of about 80 kBq, and 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 a dose of some concern from secondary contamination appeared unlikely, but could not be excluded.

The source material, with which Mr Litvinenko was allegedly poisoned, could however have been a much greater potential hazard to people than that posed by secondary contamination from the victim. 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 INDIVIDUAL MONITORING FOR POLONIUM-210

Since environmental (surface contamination) monitoring of places associated with the incident did not start until a few weeks after the date of the presumed poisoning, such monitoring provided information on the remaining hazard, from which advice on the need for any remediation measures could be given, but gave only limited information on the original extent of contamination, because of activities such as cleaning that took place in the intervening time.

In order to confirm that doses from secondary contamination from the victim (and perhaps other persons) were low, and to determine any exposures resulting from inadvertent exposure to the source material, consideration was given to the possibilities for individual monitoring for ^{210}Po. This would enable a direct assessment of any exposures that had occurred and allow the resulting individual risks to be determined.

Ideally, direct measurements of the ^{210}Po present in the body would be made using *in vivo* measurements with external detectors, usually known as “whole-body monitoring”.

However, because of the low level of photon emissions from ^{210}Po, a large amount has to be present to detect and measure it by gamma-ray spectrometry. It is estimated that the RPD low-background facility could detect about 2 MBq in the body in a 45-minute measurement. This would correspond to an intake of about 20 MBq and resulting effective dose of about 5 Sv. Hence, while *in vivo* measurements could be used to confirm a large intake, the absence of detectable ^{210}Po would not provide reassurance.

As described above and shown in Figure 1, there is ongoing excretion of about 1.5% per day of the systemic activity, so consideration was given to measurements of excreta.

Measurements of faeces have two advantages over urine measurements in the case of ^{210}Po, because it is the predominant excretion route:

- there is more activity present in a daily sample and so the sensitivity is greater;
- uncertainty in the urine: faecal excretion ratio in the model has less influence on the uncertainty in the dose assessed from the sample activity.

However, the latter is offset by the greater day-to-day variation in faecal excretion of radionuclides than in urinary excretion that is generally recognised: in some workplaces 3-day faecal samples are obtained to take account of this. Furthermore, faecal excretion of dietary ^{210}Po is likely to be higher and more variable than that of urinary excretion of ^{210}Po. Measurements of urine have distinct practical advantages over measurements of faeces in sample collection and acceptability, containment, and laboratory analysis. Provision of samples by members of the public was voluntary, and many more would be willing to provide urine than faecal samples. Urine sampling has been used extensively for monitoring of workers potentially exposed to ^{210}Po (Leggett and Eckerman, 2001). In addition, polonium is a relatively volatile element. The determination of polonium in a matrix of organic matter requires digestion in acid at low temperatures, rather than high temperature ashing in a furnace. Of these two forms of excreta, urine is much more amenable to acid digestion.

The decision was therefore made to initiate a monitoring programme based on the collection and measurement of urine samples. Consideration therefore had to be given to the range of natural levels of ^{210}Po in urine, and the detection limit that could be achieved in routine measurements. These factors determine the minimum detectable dose that could be expected from urine measurements. This would indicate the extent to which individual monitoring could provide confirmation that exposures were low, as well as enabling any exposures of concern to health to be identified.

2.1 Natural levels of polonium-210 in urine

Polonium-210 is present in environmental foodstuffs, drinking water and in the air. It is also formed within the body as a result of intakes of lead-210 (^{210}Pb), which decays to ^{210}Po. An initial search of the literature was carried out for the results of measurements of 'natural' levels of ^{210}Po in human urine, i.e., those from dietary and other intakes of ^{210}Pb and ^{210}Po (e.g. cigarette smoke), taking account of results in which levels were considered to be enhanced for some reason, such as unusually high intakes because of concentration in particular foods, or areas of high natural background. The results of this

initial survey (carried out in the first day or so following the identification of ^{210}Po in Mr Litvinenko) are summarised in Table 1. Most results in Table 1 fall in the range 5 – 15 mBq per day, with higher levels within this range being found in smokers. It was therefore considered that results above 30 mBq were very unlikely to be entirely due to 'natural' excretion.

The collection of information on natural levels of ^{210}Po in urine has continued. For some studies, individual results have been reported, allowing distributions of measured values to be compiled. Two hundred and twenty such measurements were obtained from published reports (Okabayashi et al 1975, Spencer et al 1977, Clemente et al 1980, Azeredo and Lipsztein 1991, Mancini et al 1984, Fenzi 1986, Hunt et al 1993, Santos et al 1994, 1995 and Naumann et al 1998) and a further 10 individual measurements, not yet published, received by private communication (Hunt 2007). The mean value for the individual measurements was 17 mBq d^{-1} (SD 17, range 1.2 – 111 mBq d^{-1}) with less than 12% of the measurements greater than 30 mBq d^{-1}.

However, on inspection of the data it became apparent that there were at least two distributions present: one for studies carried out in Italy (n=104) and another for the remainder of the data (n=126). The Italian and "other" results are shown as open and solid bars, respectively, in Figure 3. The mean value of the Italian data is 26 mBq d^{-1} (SD 20, range 6 – 111 mBq d^{-1}), with about 25% of the results above 30 mBq d^{-1}. In contrast, the mean value of the "other" measurements is lower at 9.0 mBq d^{-1} (SD 6.9, range 1.2 – 57 mBq d^{-1}), with less than 1% of the results above 30 mBq d^{-1}, supporting the original assessment. For some groups of UK residents (Section 5) and for overseas visitors not considered to be at high risk of exposure (Section 6) 4% or less of results were above 30 mBq d^{-1}, clearly inconsistent with the Italian studies, but not with the "others".

Table 1 Reported values of ^{210}Po excretion in urine

Country	Subjects	Range	Reference
USA		9.3 mBq d^{-1}	Spencer et al., 1977
Germany		Detection limit (2 mBq l^{-1}) to 9.9 mBq d^{-1}	Naumann et al, 1998
Brazil	Non-smokers	5.2 +/- 2.2 mBq d^{-1}	Lipsztein et al, 2003
	Smokers	9.9 +/- 4.1 mBq d^{-1}	
Saudi Arabia	Non-smokers	1.5 to 10 mBq l^{-1}	Al-Arifi et al, 2006
	Smokers	3.3 to 15.9 mBq l^{-1}	
	Shisa smokers	2.2 to 19.6 mBq l^{-1}	

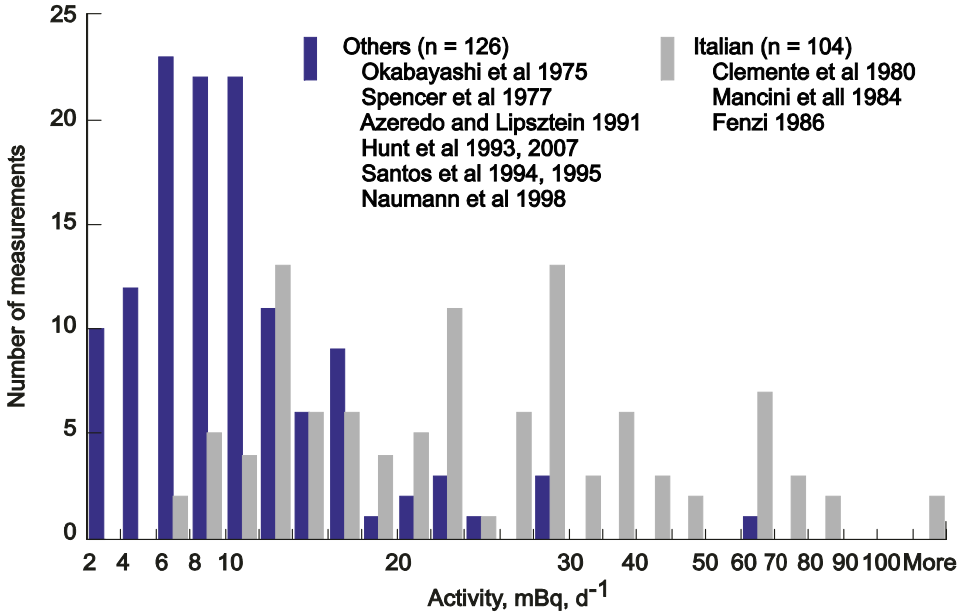

Figure 3 Reported Naturally Occurring Levels of ^{210}Po in Urine

2.2 Measurement of polonium-210 in urine

Different methods can be used for determination of the ^{210}Po in a sample according to the objectives of the measurement. There are several different analytical techniques available, which are complementary, and fit for their respective purposes. Generally the greater the precision and sensitivity needed, the longer the process will take (a) because of the need for chemical processing to isolate the polonium, and (b) to allow time for a sufficient number of radioactive decays to take place.

The Health Protection Agency measurements were designed for public health purposes: primarily to confirm that exposures were low for members of the public and various employees who may have had contact with a contaminated person or location, and to confirm that none had acquired significant amounts that could have adverse heath effects, or require long-term biological monitoring. A sensitive, but relatively rapid, method was developed for the incident by RPD staff (Ham, 2009). It requires 2–3 days from receipt of a 24-hour urine sample. The method was adapted from one that is in routine operational use for measurements on environmental samples, e.g. food. It is therefore capable of measuring natural levels of ^{210}Po in many types of sample, including urine. It is summarised in Figure 4.

During planning it was estimated that a Minimum Detectable Activity (MDA)* of 20 mBq per day could be expected routinely. In practice MDAs of 1–10 mBq per day were

* For the circumstances of a particular measurement, the MDA is the activity that, if present in the sample, would be 95% certain of detection. Thus, if a very large number of samples were measured, each containing an activity equal to the MDA, activity would be detected in 95% of them.

achieved, depending on the recovery, i.e., the fraction of polonium present in the sample that is deposited on the silver disc. The recovery, and hence the MDA for each measurement, was determined by measuring the amount of yield tracer deposited on the disc. The method is remarkably sensitive: an activity of 1 mBq ^{210}Po corresponds to only 6 x 10^{-18} gram polonium.

During validation of the procedure, measurements were made on urine samples from RPD staff. These confirmed that the procedure was able to measure natural ^{210}Po levels in both smokers and non-smokers. Validation checks were also carried out 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. Nevertheless the results obtained have all been consistent, which gives further confidence in the reliability of the data.

Consideration of the distribution of natural background levels and the sensitivity of the measurement techniques resulted in the choice of a value of 30 mBq d^{-1} for the “Reporting Level" (RL), above which a dose calculation would be performed. This RL corresponds to a minimum detectable committed effective dose E(50) of about 0.3 mSv (for a urine sample obtained 20 days after intake) based on the cautious assumption of 100% intake by inhalation (see Section 4.3 below). The fraction of the intake excreted in urine per day decreases with time after intake, as the activity remaining in the body decreases. Hence the intake calculated to give a certain activity excreted per day increases as the time between intake and sample collection increases, and with it the dose assessed from the intake. Thus the dose assessed from 30 mBq d^{-1} increased to 1 mSv for a sample provided about 100 days after the presumed intake (typically February 2007). Thus it was feasible to confirm that doses were well below levels considered to be of possible health concern, even for urine samples collected several months after intake.

1. Measure 1 litre of urine sample using a measuring cylinder into a 2-litre beaker.

2. Add ^{209}Po or ^{208}Po yield tracer (typically about 0.2 Bq)

3. Add 200 ml of concentrated nitric acid. Heat on a hotplate set at 200°C with occasional stirring. The sample should go straw coloured over time.

4. Evaporate sample to dryness (overnight on a hotplate set at 150°C).

5. Cover the residue with the minimum quantity of concentrated hydrochloric acid (enough to dissolve the residue with warming) and take to dryness (hotplate 200°C). Repeat this step.

6. Dissolve the residue with 6M hydrochloric acid and transfer to a suitable beaker (250 – 600 ml tall form beaker) using 6M hydrochloric acid. Make up to half beaker volume using 6M hydrochloric acid.

7. Add 1 ml of 30% w/v hydroxyl ammonium chloride solution. Add a magnetic stirrer and adjust the pH of the solution with stirring to 2, with 0.880 ammonia or hydrochloric acid.

8. Heat the solution with stirring to at least 85°C. Meanwhile put a clean silver disc into a disposable holder such that only one side of the disc is exposed. Place the holder containing the silver disc in the solution. Under these conditions polonium will electrodeposit spontaneously onto the silver disc. After 3 hours remove the holder from the solution, take out the disc, rinse and allow it to dry.

9. Count overnight on a solid state alpha spectrometer: 12 hours should be long enough to measure a minimum detectable activity (MDA) of 20 mBq for a 24-hour sample.

Figure 4 Summary of procedure used to measure ^{210}Po activity in a urine sample

3 DEVELOPMENT OF THE INDIVIDUAL MONITORING PROGRAMME

3.1 Overall strategy and uses of the individual monitoring programme

It was apparent early in the incident that the programme would have to be able to deal with large numbers of persons and urine samples. As a result of the identification by the police of several contaminated locations within the first few days, the number of persons requiring assessment rapidly expanded to include several hundreds. The strategy developed by staff from the Agency's Local and Regional Services (LaRS) and the RPD was therefore to obtain samples from those with the highest assessed risk of exposure to environmental ^{210}Po at each location, in order to assess whether these individuals had intakes of concern for health effects. In addition, the results would provide important information on whether more people at the site needed to be monitored, or whether reassurance could be provided to others associated with the site but considered to be at a lower risk of exposure.

3.2 Public health risk assessment and selection of persons for urine monitoring for polonium-210

Each contaminated site identified by the police was surveyed for contamination with ^{210}Po by RPD staff. (Information on the extent of ^{210}Po contamination measured during forensic investigations was provided by the organisation whose staff carried out the initial environmental monitoring of possible crime scenes in its role of advising the Metropolitan Police.) Where these surveys established a degree of environmental contamination sufficient to pose a potential risk of contamination of persons associated with the site, a public health risk assessment was undertaken by medical public health consultants from the Agency's LaRS Division.

LaRS consultants reviewed the environmental findings and conferred with management and senior staff at the affected sites to determine the range of persons potentially at risk of internal contamination with ^{210}Po. The potential sources and pathways for transmission of the environmental contamination were considered, together with the nature of the activities undertaken by persons at the affected site, through their occupation or connection with the site as clients, customers, guests or visitors.

All persons identified as potentially at risk of contamination with ^{210}Po were administered a face-to-face questionnaire designed to characterise activities and behaviours at that site that could be associated with risk of internal contamination. Questionnaires were developed from common templates and adapted and extended by public health consultants to encompass site-specific risks. All persons were asked about smoking status, and women were asked whether they were pregnant. Questionnaires were used to inform assumptions by RPD staff about possible routes of exposure as part of dose assessment (see Section 4.4). Selected questions from the questionnaires are given in Figures 5 and 6.

In a minority of cases, the questionnaire enabled persons to be excluded from the need for urine monitoring. At the hospital sites, the questionnaires were used to identify a subgroup of persons to be invited to submit urine samples, comprising staff involved directly in Mr Litvinenko's patient care and who reported direct contact between the patient's body fluids and their personal protective equipment or skin. At one hotel, initial monitoring was confined to bar staff and staff servicing/cleaning contaminated guest rooms and public areas; additional staff were assessed and monitored after a high prevalence of internal contamination was found among bar staff. At most sites however, all persons initially identified as potentially at risk were invited to submit 24-hour urine samples for measurement of ^{210}Po activity.

Questionnaires were subsequently used, together with the results of urine tests, to assess the occupations, activities and behaviours associated with the risk of personal contamination with ^{210}Po for persons in these contaminated locations.

A significant number of persons needing urine testing were identified through callers to NHS Direct (NHSD), the UK national healthcare telephone advice service. NHSD administered standard questionnaires to callers relating to the incident. A substantial number of persons who were customers at specified sites on certain dates were identified and offered testing. These questionnaires were also used to exclude callers not in this risk category.

LaRS staff arranged collection of urine samples from individuals identified as eligible for testing. RPD staff organised transfer from a central collection point in London to the Centre for Radiation, Chemical and Environmental Hazards (CRCE) at Chilton in Oxfordshire, measured the ^{210}Po activity in the samples (or arranged its measurement at another laboratory), assessed doses from them (see Section 4) and reported the results back to LaRS.

Once measurement and dose assessment were completed, LaRS staff communicated the result to the individual. Up to three attempts were made to contact people by telephone to communicate results below 6 mSv, described as being 'of no health concern' (Categories 1, 2 and 3a, see Section 4). Telephone communications were followed up with a confirmatory letter. For individuals with assessed doses of ≥ 6mSv (Category 3b), arrangements were made to give the results in a face-to-face interview with a senior public health doctor who was able to explain the possible health consequences. These people were also offered the opportunity to have their homes monitored to provide further reassurance that they had not tracked radioactivity from contaminated sites, and follow-up urine measurements.

1. Were you involved in the direct care of the patient?
2. What is your occupation (nurse, doctor, radiology staff, domestic worker etc)?
3. What is your location (ward, A&E, ICU, laboratory, imaging etc)
4. Did you or your protective personal equipment (PPE) come into direct contact with urine, faeces, vomit, blood, or other body fluids of the patient?
5. Were there occasions when you did not wear the standard personal protective equipment for the work you undertook?
6. Were there occasions when you did not follow the prescribed hygiene rules?

Figure 5 Some questions asked of selected Health Care staff, who had worked in circumstances with potential exposure to ^{210}Po. All answering "yes" to Questions 1 and 4 were asked to provide a urine sample

1. Did you work in the hotel on any day between aa/bb/2006 and xx/yy/2006 ?
2. What is your designation (eg bar staff, porter, housekeeping, waiter room service, etc)?
3. Did you work in any of the following rooms (Room AAA, Room BBB, Room CCC)? If YES, what did you do (carrying bags, cleaning bathrooms, and toilets, room service etc)
4. Did you work in, or have any exposure to Bar Y of the Hotel?
5. Have you been ill? If YES, have you had any of the following: Nausea, vomiting, diarrhoea, fever, sore throat, bleeding gums, unusual bleeding from cuts.

Figure 6 Some of the questions asked of selected Hotel staff

3.3 Practicalities of urine sampling for large numbers of people

The methodology for measurement of ^{210}Po activity in a urine sample (and assessment of the dose from it, see below) could be readily adapted from existing procedures, since staff were available with expertise in developing methods, not only in applying established ones. However, the requirement to process large numbers of samples, and to report results rapidly to inform the need for further measures and/or provide reassurance, posed some specific challenges including:

- availability of urine sample bottles in sufficient quantities;
- the need for clear instructions on how to provide 24-hour samples (for people whose first language might not be English);
- organisation of transport arrangements at short notice;
- laboratory sample throughput and analysis time – potentially for hundreds, if not thousands, of samples;
- the need for assistance from other UK laboratories with sample measurements;
- the provision for international assistance with sample measurements if needed;
- storage and disposal of urine samples.

When it became apparent that numbers could exceed the RPD's analytical capacity of about 40 samples per day, RPD staff contacted other laboratories in the UK that carry out low-level measurements of ^{210}Po in environmental samples. Some offered support, which meant rearranging their existing programmes and dedicating laboratory space and equipment to this work. They were sent check samples to confirm consistency of results, before being sent any real samples. As a back-up, several laboratories in Europe were also contacted and sent check samples. Although about 500 samples were received within the first month (Figure 7), all were analysed in the UK. However, a number of European laboratories analysed samples from their own citizens. Samples from a total of 753 people were processed in all: another 250 up to the end of May 2007, and one in each of June, July and August 2007.

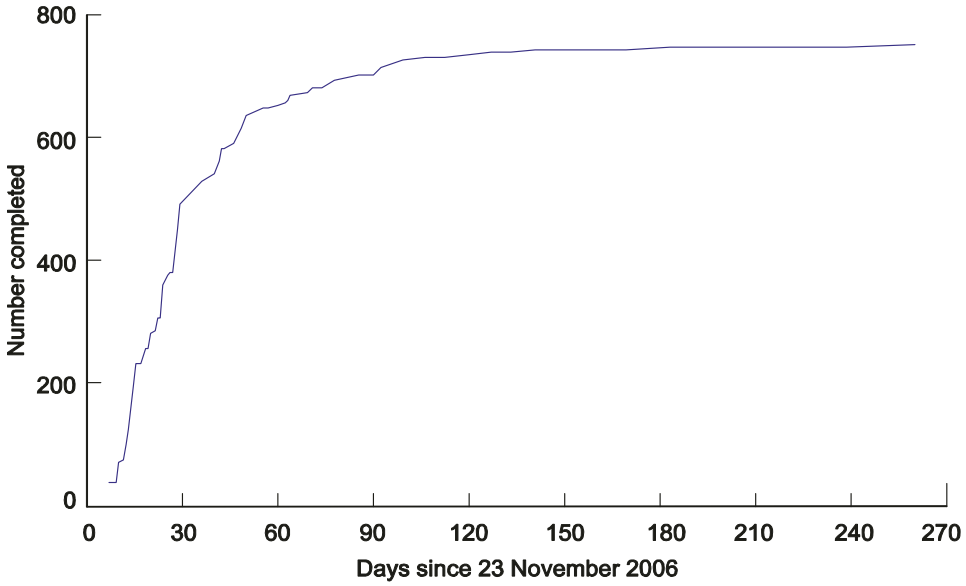

Figure 7 Throughput of samples and dose assessments up to 10 August 2007

3.4 Recording of results of measurements and dose assessments

Information relating to each urine sample came from four sources:

- extracts from the LaRS risk assessment questionnaire, (Section 3.2) including smoking status, date of birth, and (where known) date of initial possible exposure and possible exposure duration (the entire questionnaire where a special dose assessment was required, Section 4.4);
- a sample record form, completed by the individual;
- the urine measurement report form, provided by the Radiochemistry Team;
- the dose assessment report forms, provided by the Dose Assessments Group.

3.4.1 The sample record form

This form accompanied the empty 2.5-litre urine sample bottles that were despatched from RPD to LaRS staff. It was used to record the name, address, occupation, place of employment and telephone number of the individual providing the sample, and an identifying bottle number. The individual was asked to provide a 24-hour sample, and to give the start date and time, and the end date and time of the sample. The form also explained the need to bath or shower before the first collection, how to collect a 24-hour sample of urine, and how to return it. LaRS staff informed the individual about when and how they could expect to receive the results, and gave brief information on the radiological hazard presented by ^{210}Po. When the urine sample was returned to RPD, a Unique Identifier (UID) in the format RI 06/xyz was added to the form and written on the bottle (or bottles).

3.4.2 The urine measurement report form

This form was used to record the activity concentration in urine measured by alpha spectrometry, in units of Bq per litre. It was also used to record the UID, the activity reference date, the mass of the sample, the total sample activity (Bq) the volume analysed and the sample duration (normally 24 hours).

3.4.3 The dose assessment report forms

In most cases, a 'standard' dose assessment was all that was required (Section 4.3). The corresponding report form recorded the UID, the sample end date, the mass of the sample, the duration of the sample, the activity present in the sample, the activity in a 24-hour sample (inferred if the duration was less then 24 hours), and the assessed dose. If a special dose assessment was required (Section 4.4), then the assessed dose was recorded on an individual report form.

3.4.4 Individual monitoring results database

A robust and reliable system was needed to bring together all the information on each sample, to ensure that the correct result was returned to the individual who provided it. This had to be done in a way that would ensure confidentiality of information that could identify an individual. This system had to be developed and tested in parallel with processing the first samples, the results of which were required urgently to determine whether further measures were needed. Initially an EXCEL spreadsheet was produced for expediency, but it was in due course replaced by an ACCESS database.

The database was used to provide regular "medical-in-confidence" dose reports, usually on a daily basis. These reports contained the UID, the name of the individual, the date of birth (as a secondary means of identification), the "site of contamination", the date of receipt of the urine sample, the activity in the 24-hour urine sample, and the assessed dose. The "site of contamination" field was used to identify the category of potentially-exposed individuals to which an individual was allocated (e.g. guests at a particular hotel, health care workers at one of the hospitals). Measured activities were reported either as "BRL" (below reporting level) or as the measured value in Bq d^{-1}. For BRL results, no dose assessment was reported. Otherwise, the dose was reported either as the assessed value in mSv, or as "less than 1 mSv". These dose reports were the primary means for reporting results back to individuals by LaRS staff.

Another requirement was to provide frequent (initially, daily) up-to-date summaries of the programme status and results to the RPD Operations Team at CRCE, Chilton, and the National Emergency Co-ordination Centre (NECC) at the Health Protection Agency's London Headquarters. These were merged with data on identification of at-risk individuals and sample requests held by the LaRS London Regional Epidemiology Unit to provide regular briefings to the NECC, Cabinet Office and the media.

4 DOSE ASSESSMENT

"Dose assessment" is the process by which intakes of radionuclides, and the resulting doses, are assessed from measurements of the amount of activity present in the body (or in specific organs) and/or measurements of the amounts in samples of excreta. The dose assessment methods generally adopted for use around the world are those developed by the ICRP over the last half-century or so (e.g. TGLD 1966, ICRP 1979, 1993, 1994a, 1994b, 1995, 1998, 2006).

4.1 Stages in dose assessment process

For an individual dose assessment based on one or more measurements, the process consists of two basic stages, (i) assessment of the intake (ii) assessment of the dose resulting from that intake. These involve several steps, and the process may be summarised as follows:

1. For a given intake (typically "unit" intake of 1 Bq) by the appropriate route (inhalation or ingestion), use the ICRP biokinetic models to calculate the measured quantity (in this case the daily excretion of ^{210}Po in urine), as a function of time after intake.
2. Using that function, calculate the intake that is predicted to give the measured value at the time of measurement.
3. For that intake, calculate the amount of ^{210}Po in each body tissue as a function of time.
4. Calculate the number of radioactive decays that take place in each tissue.
5. Calculate the committed "absorbed dose" to each tissue, i.e., the energy (in the form of ionising radiation) deposited per unit mass of tissue as a result of the decays. By convention, committed doses are usually calculated for 50 years after intake. However, in the case of ^{210}Po, because the physical and biological half-lives are short, nearly all the committed dose is received within a few months of the intake.
6. Calculate the committed equivalent dose to each tissue by multiplying by the radiation weighting factor.
7. Calculate the committed "effective dose", the sum of the tissue equivalent doses each multiplied by the corresponding tissue weighting factor, which gives a broad estimate of the risk of a fatal cancer, which is typically taken to be about 0.005% per mSv (ICRP 1991).

The process, as applied to measurements made in the polonium-210 incident, is considered in more detail in the following Sub-Sections (4.1.1 – 4.1.3). These are followed in Sections 4.2 – 4.4 by a description of the procedures that were used.

Except where stated otherwise (Table 5), all calculations described below and carried out by the Health Protection Agency in the monitoring programme used the current

ICRP biokinetic and dosimetric models as applied in ICRP Publication 68 (ICRP 1994a). These models form the basis of formal dose assessments currently undertaken in the UK, *i.e.*, the ICRP Publication 67 systemic model for polonium (ICRP 1993), the ICRP Publication 66 Human Respiratory Tract Model (HRTM, ICRP, 1994b), and the ICRP Publication 30 model for the GI tract (ICRP, 1979). For ingestion, the fractional absorption in the GI tract (f_1 value) is assumed to be 10%. For inhalation, absorption Type M is assumed, *i.e.*, 10% of the deposited material dissolves rapidly and is absorbed into blood at a rate of 100 d^{-1}, and the remaining 90% dissolves at a rate of 0.005 d^{-1}. An f_1 value of 10% is also assumed for material cleared to the throat and swallowed (see Section 4.4.3).

In Sub-Sections 4.1.1 – 4.1.3, calculations are based on a reference worker (adult male undertaking light work, a mixture of sitting at rest and light exercise), but similar results would be obtained for other subjects under exposure conditions likely to have been relevant to this incident.

4.1.1 Urinary excretion following intake of ^{210}Po

Figure 8 shows urinary excretion (mBq d^{-1}) following an intake of 1 kBq by ingestion or inhalation. These units were chosen for clarity, as representative of values obtained in the monitoring programme (rather than simply giving the conventional fractional rates in Bq d^{-1} following an intake of 1 Bq).

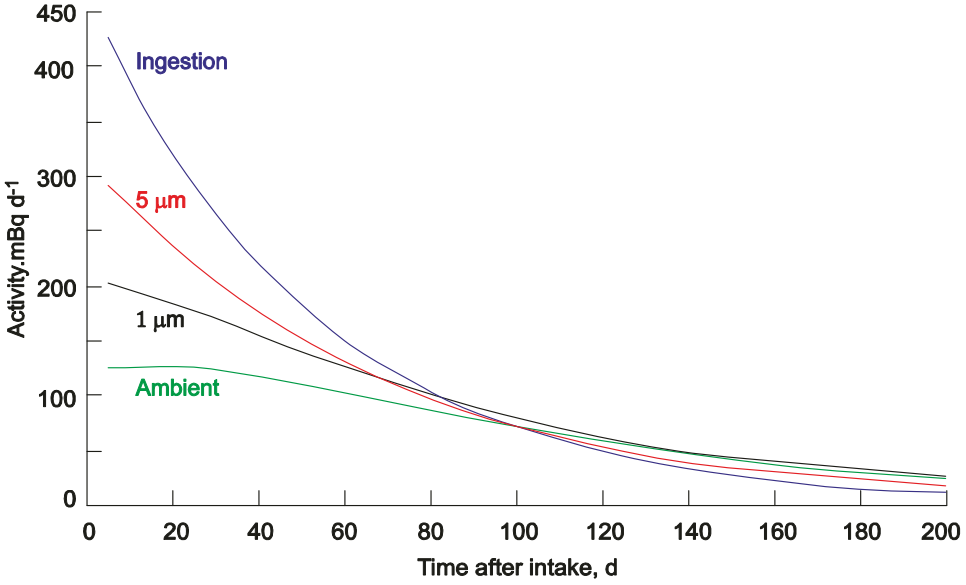

Figure 8 Urinary excretion following intake of 1 kBq ^{210}Po by ingestion or by inhalation of 5-µm AMAD, 1-µm AMAD or "ambient" aerosols

To see how these results arise, consider someone who ingested 1 kBq (1000 Bq) of ^{210}Po. About 10% of the ^{210}Po 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 (Section 1.1). Initially that would result in about 0.5 Bq per day (500 mBq d^{-1}), but it decreases with time as the amount of ^{210}Po in the body decreases. So, as shown in Figure 8, by the time the first measurements were made (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}.

Inhalation is more complex. Some of the inhaled activity is exhaled again or deposits in the front of the nose, where there is assumed to be no absorption to blood, and this depends on the size of the inhaled particles. Three size distributions were considered (Section 4.4.4) and results for them are shown in Figure 8: AMAD (activity median aerodynamic diameter) of 1 μm and 5 μm, which are the ICRP default assumptions for exposure of members of the public and workers, respectively, and an "ambient" aerosol, which assumes that the ^{210}Po is attached to particles in room air in the same way as radon decay products.

Table 2 shows deposition in each respiratory tract region for these three aerosols (inhaled by a reference adult male at light work). For the 5-μm aerosol there is high deposition in the extra-thoracic airways (ET_1 and ET_2). For progressively smaller aerosols (1-μm and ambient) there is less deposition in the upper airways (ET_1, ET_2 and BB regions) and more in the lower respiratory tract (bb and AI regions).

Table 2 Percentage of inhaled material deposited in each respiratory tract region (reference worker), and resulting percentage of inhaled ^{210}Po rapidly absorbed into blood

Aerosol size	AMAD 5 μm	AMAD 1 μm	Ambient*
Percentage of inhaled material deposited			
Region			
ET_1 (anterior nasal passage)	33.9	16.5	5.9
ET_2 (posterior nasal and oral passages, pharynx and larynx)	39.9	21.1	6.8
BB (bronchial: trachea and airway generations 1–8)	1.8	1.2	0.66
bb (bronchiolar: airway generations 9–15)	1.1	1.7	2.5
AI (Alveolar-interstitial: airway generations 16 and beyond, the gas-exchange region)	5.3	10.7	11.7
Total excluding ET_1 (ET_2 + BB + bb +AI)	48.1	34.7	21.7
Percentage of ^{210}Po intake absorbed into blood by 10 days	6.8	4.9	3.1

* Ambient: Trimodal distribution typical of room air as defined in Table 4

It is assumed in the HRTM that material deposited in the front of the nose (ET_1) is removed by nose blowing, and that activity is not absorbed from it to the blood. Hence the fraction of the intake absorbed rapidly, and so appearing in urine initially, depends on deposition in the other regions. As shown in Table 2, this decreases from about 50% of the inhaled activity for the 5-μm aerosol, to about 20% for the ambient aerosol. The amount rapidly absorbed into blood, resulting from the rapid fraction of absorption from the respiratory tract and absorption in the GI tract of material cleared from the respiratory tract and swallowed, is about 15% of the total deposit in regions ET_2, BB, bb and AI combined.

Based on the assumptions made here, if 1 kBq was inhaled, the amount initially absorbed into blood, and hence the early excretion in urine, is 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 ^{210}Po continues to enter the blood, and urinary excretion drops more slowly than after ingestion.

4.1.2 Intake of ^{210}Po assessed from a urine measurement

As shown in Figure 8, 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 in urine at 100 days then the intake is estimated to be about 1 kBq. As shown in Figure 9 (which was derived from the results shown in Figure 8), 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.

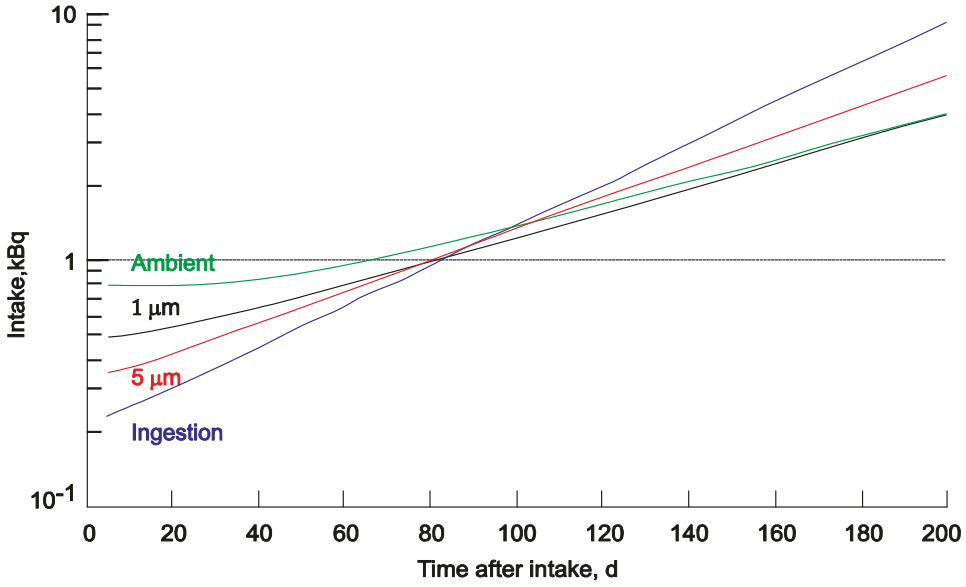

Figure 9 Intake by ingestion, or by inhalation of 5-µm AMAD, 1-µm AMAD or "ambient" aerosols, estimated from a measurement of 100 mBq d^{-1} ^{210}Po in urine

4.1.3 Dose assessed from a urine measurement

The dose coefficients (committed effective dose per unit intake) for inorganic compounds from ICRP Publication 68 (ICRP 1994a), are 2.4 x 10^{-7} Sv Bq^{-1} (ingestion), 2.2 x 10^{-6} Sv Bq^{-1} (inhalation Type M, 5-µm AMAD) and 3.0 x 10^{-6} Sv Bq^{-1} (inhalation Type M, 1-µm AMAD). Hence doses calculated for intake of 1 kBq are 0.24, 2.2 and 3.0 mSv respectively. For intake of 1 kBq ^{210}Po as an "ambient" aerosol a dose of 3.3 mSv was calculated here. Doses from inhalation are much higher than for ingestion because of the additional dose to the lungs, which are assigned a high tissue weighting factor. The committed effective dose assessed from a measurement of 100 mBq d^{-1} at any time is obtained by multiplying the intake (as shown in Figure 9) by these doses per kBq. The results are shown in Figure 10. 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 an

order of magnitude lower. Figure 10 also shows results assuming that intake was a mixture of ingestion and inhalation (in this case 50% inhalation with a 1-µm AMAD), because a mixture was assumed in most special assessments (see Section 4.4.6). For a measurement different from 100 mBq d^{-1}, the result can simply be scaled in proportion.

As shown in Figure 10, 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.

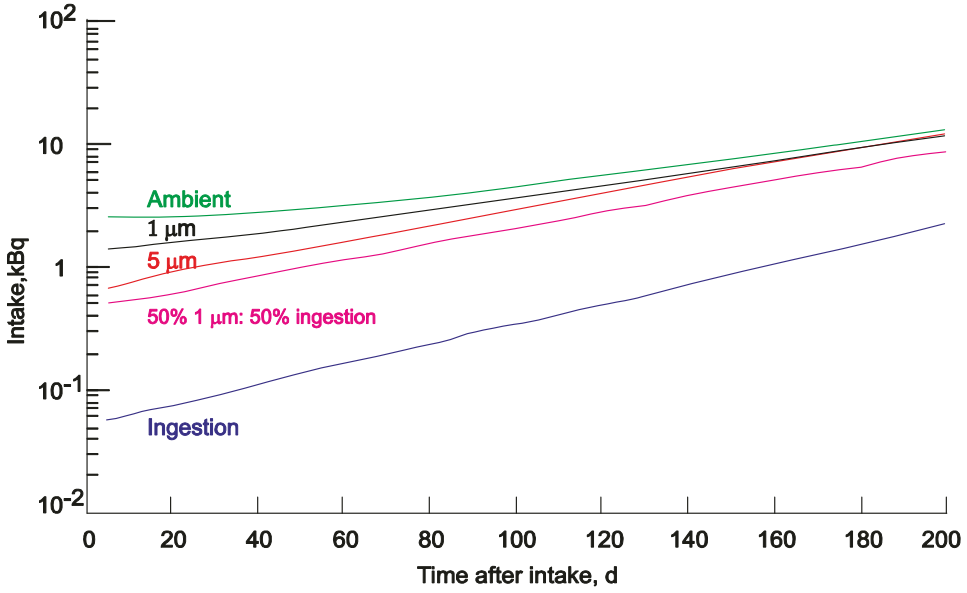

Figure 10 Dose assessed from a measurement of 100 mBq d^{-1} ^{210}Po in urine, for intake by ingestion, by inhalation of 5-µm AMAD, 1-µm AMAD or "ambient" aerosols, or from a mixture (50% inhalation of 1-µm aerosol and 50% ingestion)

4.2 Dose assessment procedure for response to the polonium-210 incident

It was anticipated that it could be necessary to perform assessments on large numbers of people, and that results would be required promptly to guide the need for further measures and for reassurance, as noted above. A categorisation system was therefore developed by which rapid assessments would be made for those individuals whose urine measurements indicated that their intakes and doses were trivial, while thorough assessments would be made for those individuals likely to have received greater intakes and doses. This is summarised in Figure 11. The procedure is outlined immediately below and described in detail in the following Sections (4.3 and 4.4).

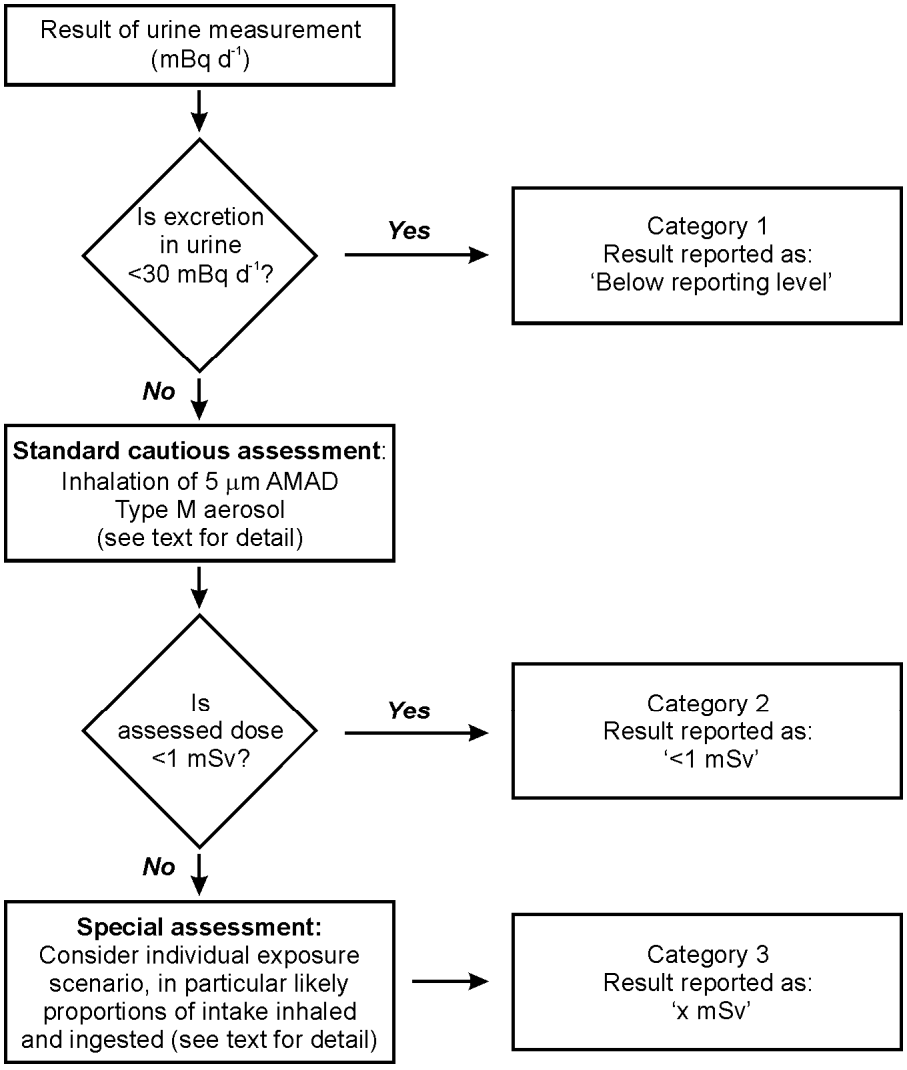

Figure 11 Procedure for assessment of doses from ^{210}Po in urine samples

In the first stage, the measurement result was compared with a "Reporting Level" (RL), set at 30 mBq d^{-1} (see Section 2.2 above). It was considered that measurements above this level would probably include a contribution from contamination from the incident. Measurements below the RL were assigned to Category 1. The second stage, applied to measurements above the RL, was to carry out a simple 'standard' assessment. This used assumptions expected to overestimate doses in most cases, notably that of 100% intake by inhalation. If the standard assessment gave a dose of less than 1 mSv, the result was assigned to Category 2, and reported as simply "<1 mSv". These two stages are described in Section 4.3.

If the standard assessment gave a dose of more than 1 mSv, the result was assigned to Category 3 and a more detailed 'special' assessment was carried out (Section 4.4). As described below, doses assessed to be <6 mSv (Category 3a as well as 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".

Note that this dose assessment protocol had to be defined on a short time scale (about 48 hours) because the first urine sample measurements became available within a few days of the start of the programme and results were required promptly. At that time, information on the circumstances of possible exposures was, of course, sparse.

4.2.1 Choice of dose categories

Pragmatic decisions had to be made, again in a short period of time, on how to categorise the doses. The criteria underpinning the dose categories and their basis are described below. However it is emphasised that other criteria could have been justified and that in any future radiation incident different criteria may be appropriate, depending on the specific circumstances such as the number of people being measured.

4.2.1.1 1 mSv Criterion

The choice of 1 mSv as a criterion for conducting special assessments and reporting the assessed doses in full, was based on various considerations (HPA 2007a, 2007c). In particular the annual dose limit for members of the public for radiation exposures resulting from situations subject to regulatory control (IRR 1999) is 1 mSv. This dose limit does not apply in the case of the London poisoning incident as the presence of polonium-210 was as a result of malevolent actions and not as a result of a controlled situation. Nevertheless the dose limit of 1 mSv for controlled situations does provide a perspective on this level of dose to which members of the public could readily relate. The polonium-210 exposure is considered to be a single incident, and so the risk is considerably lower than that from exposure at 1 mSv every year. Hence on that basis a higher value could have been justified here. A dose of 1 mSv is also about 50% of the typical annual dose from natural background radiation. Individual dose assessments are not normally carried out for exposures of the public at such levels.

4.2.1.2 6 mSv Criterion

It was considered that there was a need to have a further categorisation, both in terms of presenting the significance of the doses, and in identifying a group of people to whom further measurements would be offered as reassurance that the radioactivity in their body was decreasing with time as predicted by existing knowledge of the behaviour of polonium in the body (Section 1.1). In reporting results to those who provided the samples, the media, and others without radiation protection expertise, emphasis was placed on giving a clear and simple message. This was difficult, because it is assumed for radiation protection purposes that there is no threshold for radiation induced cancer: any radiation exposure carries some additional risk, although the risk decreases with decreasing dose. With this in mind doses assessed to be <6 mSv were described as being “of no concern”, and doses assessed to be ≥ 6 mSv were described as being “of some concern” (HPA 2007a, 2007c). As for the 1 mSv criterion, the value of 6 mSv was based on several considerations, in particular it was noted that where a worker is routinely exposed to ionising radiation (IRR 1999), medical surveillance would be required where exposures exceed 3/10th the dose limit of 20 mSv in a year (i.e. 6 mSv). Another relevant consideration is that the UK Action Level for radon in homes is set at a concentration of 200 Bq m^{-3}. This corresponds to an effective dose per year of about 10 mSv, most of which results from alpha-irradiation of the lungs by the short-lived radon decay products ^{218}Po and ^{214}Po. Home owners are advised to take action to

reduce the radon concentration if it is above the Action Level, which thus represents a level of “some concern”, but again applied to continuous, rather than one-time exposure. Lifetime effective doses from radon in dwellings above the Action Level are likely to be hundreds of mSv or more.

For doses below 6 mSv, from the risk factors given by ICRP in Publication 60 (1991), any increase in the risk of cancer, which is the main concern, will be less than about 0.03%.

4.3 Initial stages (Categories 1 and 2)

The aim of the initial stages was to divide individuals into:

- those for whom the intake, if any, was negligible, at least in the first few months of the incident (Category 1);
- those who probably had detectable ^{210}Po from the incident, but whose dose was unlikely to exceed 1 mSv (Category 2);
- those who may have received a dose greater than 1 mSv (Category 3), and for whom a more detailed special assessment would be carried out.

4.3.1 Category 1: Urine measurement < 30 mBq d^{-1}

As described in Section 2.1 above, from reports in the literature, natural background excretion of ^{210}Po is expected to be less than 20 mBq d^{-1} in most people. It was estimated that a minimum detectable activity (MDA) of 20 mBq d^{-1} could be obtained routinely, i.e., during processing of large numbers of samples, without taking special measures. It was therefore decided that those 24-hour samples for which the measured activity fell below 30 mBq should be assigned to the lowest category (Category 1). Results in this category were recorded as "Below Reporting Level" (BRL) i.e. no dose calculated or reported. Assignment to Category 1 did not necessarily mean that the individual did not have any intake of ^{210}Po from the incident. However, for a measurement made at the end of November 2006, it does mean that it is unlikely that the person had received an intake giving a dose of more than 0.3 mSv. Figure 12 shows doses assessed from a measurement of 30 mBq d^{-1} obtained by scaling the results in Figure 10. Even for measurements made after 3 months after the presumed date of intake (typically February 2007) a measurement below 30 mBq d^{-1} means that it is unlikely that the person received a dose of more than 1 mSv.

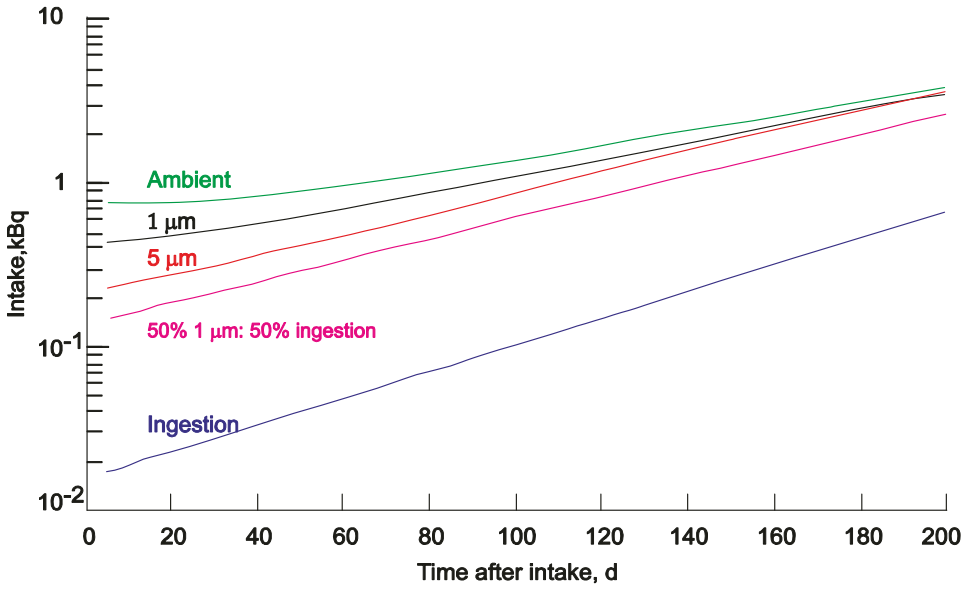

Figure 12 Dose assessed from a measurement of 30 mBq d^{-1} ^{210}Po in urine, for intake by ingestion, by inhalation of 5-µm AMAD, 1-µm AMAD or "ambient" aerosols, or from a mixture (50% inhalation of 1-µm aerosol and 50% ingestion).

4.3.2 Category 2: Urine measurement > 30 mBq d^{-1}

An initial rapid assessment was made using a standard set of assumptions, including 100% intake by inhalation. Each was chosen to be either realistic or pessimistic, i.e. to be likely to overestimate the actual dose (see list below). The assumptions made were:

- Route of intake entirely through inhalation, which gives much higher assessed doses than ingestion, because of the additional dose to the lungs – see Figures 10 and 12 (pessimistic).
- Aerosol size represented by an AMAD of 5 µm (realistic), the default assumption for occupational exposure.
- Absorption Type M (moderate) (realistic). ICRP Publication 71 (1995) recommends the use of default Type M in the absence of specific information (see Section 4.4.3).
- Fractional absorption in the GI tract (f_1) of 0.1, the default value for occupational exposure as in ICRP Publication 68 (realistic) (see Section 4.4.3). Moreover, the dose assessed from a urine sample is not particularly sensitive to the f_1 value.
- No background subtraction (pessimistic).
- Acute intake on a fixed date (realistic). Moreover, the dose assessed from a urine sample is not particularly sensitive to the assumed time of intake (Figure 12 and Table 3).

Doses were calculated by simply multiplying the sample activity by a constant (K-factor), which was calculated using these assumed parameter values. The 'K-factor' (dose per unit daily excretion, Table 3) was obtained by dividing the dose coefficient (effective

dose per unit intake) by the predicted daily excretion per unit intake. It is proportional to the curves for 5-µm AMAD aerosols in Figures 10 and 12. Hence:

Dose (mSv) = K x measurement in a 24-hour sample (mBq)

For example, if 50 mBq d^{-1} was measured in a urine sample taken 41 days after presumed intake, the assessed dose would have been 1.23 x 10^{-2} x 50 = 0.6 mSv. Since this is <1 mSv, the result would have been reported as "<1 mSv" (Category 2). For another example, if 105 mBq d^{-1} was measured in a urine sample taken 36 days after presumed intake, the assessed dose would have been 1.14 x 10^{-2} x 105 = 1.2 mSv. Since this is >1 mSv, the assessment would have been assigned to Category 3, and a special dose assessment carried out as described below.

Note that values of K are tabulated for up to 108 days. By then the value of K was 3.33 x 10^{-2} (mSv per mBq d^{-1}), and so if the measurement was >30 mBq d^{-1}, the assessed dose would have been >3.33 x 10^{-2} x 30 mSv, which is >1 mSv, and so any measurement above the Reporting Level would have been assigned to Category 3.

Table 3 Conversion factors, K, (mSv per mBq d^{-1}) for Category 2 assessments of dose from urine excretion rate, on the assumption of 100% intake by inhalation of a 5-µm AMAD Type M aerosol (see text for details)

Day*	K (x 10^{-2})	Day	K (x 10^{-2})	Day	K (x 10^{-2})	Day	K (x 10^{-2})
25	0.974	46	1.33	67	1.81	88	2.47
26	0.989	47	1.35	68	1.84	89	2.51
27	1.00	48	1.37	69	1.87	90	2.55
28	1.02	49	1.39	70	1.89	91	2.59
29	1.03	50	1.41	71	1.92	92	2.63
30	1.05	51	1.43	72	1.95	93	2.67
31	1.06	52	1.45	73	1.98	94	2.70
32	1.08	53	1.47	74	2.01	95	2.75
33	1.09	54	1.49	75	2.04	96	2.79
34	1.11	55	1.52	76	2.07	97	2.83
35	1.13	56	1.54	77	2.10	98	2.87
36	1.14	57	1.56	78	2.13	99	2.91
37	1.16	58	1.58	79	2.17	100	2.96
38	1.18	59	1.61	80	2.20	101	3.00
39	1.20	60	1.63	81	2.23	102	3.04
40	1.21	61	1.66	82	2.26	103	3.09
41	1.23	62	1.68	83	2.30	104	3.14
42	1.25	63	1.71	84	2.33	105	3.18
43	1.27	64	1.73	85	2.37	106	3.23
44	1.29	65	1.76	86	2.40	107	3.28
45	1.31	66	1.78	87	2.44	108	3.33

* After presumed intake

4.4 Special dose assessments (Category 3)

4.4.1 Introduction

4.4.1.1 Judgements

In carrying out a special assessment (Category 3), assumptions were made that were specific to the individual's exposure, about the route of intake, whether the intake was taken over a period of time, or as a single acute intake, and the aerosol size distribution. The aim was to make the dose assessment more realistic than in the standard assessment above, but nevertheless to make it unlikely that doses would be greatly underestimated.

There was often sparse information available, and so judgement was used to make decisions about the intake scenario, taking into account information available in the individual's risk assessment questionnaire. Occasionally efforts were made to obtain additional information through further enquiries.

Some parameters have little effect on the resulting dose, and so effort was focused on consideration of those that do make a difference. A sensitivity analysis demonstrated that the assessed dose is very sensitive to the route of intake (ingestion or inhalation) but relatively insensitive to the pattern of intake (acute intake or chronic over a certain period) (see Figures 10 and 12).

4.4.2 Biokinetic and dosimetric models

As in the standard assessment (Section 2.2), the biokinetic and dosimetric models from ICRP Publication 68 (ICRP 1994a) were applied. The software package IMBA Professional Plus (Integrated Modules for Bioassay Analysis, Birchall *et al*, 2007) was used in all of the special assessments, as it is designed for making such calculations. (IMBA users outside the Agency could download relevant parameter files [*.ix] from www.imbaprofessional.com.)

A comparison was made with the RPD code PLEIADES (**P**rogram for **L**in**E**ar **I**nternal **A**ge-dependent **D**os**ES**, Fell *et al* 2007), which (as well as implementing all current ICRP models) also implements the Leggett and Eckerman (2001) polonium model (Figure 2) and the new ICRP Human Alimentary Tract (HAT) model (ICRP, 2006). For ingestion, the Leggett and Eckerman model gives about twice the assessed dose for unit excretion of ^{210}Po in urine, than that calculated with the ICRP Publication 67 systemic model, while for inhalation, there is less difference (Table 5). Application of the HAT model instead of the ICRP Publication 30 GI tract model has little effect. In view of the small differences, application of these new models rather than those in current use was not justified for measurements from which the risks of adverse health effects were assessed to be small.

4.4.3 Absorption

4.4.3.1 Absorption from the respiratory tract

The ICRP Task Group on Lung Dynamics (TGLD, 1966) assigned oxides, hydroxides and nitrates of polonium to inhalation Class W, and all other compounds of the element to Class D. These classifications were adopted by ICRP in Publication 30, Part 1 (ICRP, 1979) and carried over into the assignment of compounds into HRTM absorption

Types in ICRP Publication 68 (ICRP, 1994a) thus: Type M (moderate) for oxides, hydroxides and nitrates and Type F (fast) for unspecified forms of polonium.

Information relating to inhalation of polonium compounds was reviewed in ICRP Publication 71 (ICRP, 1995), for guidance on inhalation dose coefficients for members of the public. Most of the studies reviewed indicated Type M behaviour:

- Accidental inhalation by a worker of material from an unencapsulated ^{210}Po source, probably in the form of small particles of ^{210}Po oxide (Scott and West,1975)
- ^{210}Po that condenses with cigarette smoke tar (Cohen *et al.* 1979a; 1979b)
- A sodium chloride aerosol carrying ^{210}Po as the chloride inhaled by rats (Berke and DiPasqua, 1964; Casarett, 1964)
- ^{210}Po chloride in acid solution intratracheally instilled into rats (Thomas and Stannard, 1964)
- ^{210}Po hydroxide colloid intratracheally instilled into rabbits (Morrow and Della Rosa, 1964)

The only others, measurements of the *in vitro* dissolution of radionuclides in coal fly ash (Kalkwarf et al., 1984) and calcined rock dust (Kalkwarf and Jackson, 1984) indicate Type M or S behaviour for the ^{210}Po present.

ICRP Publication 71 (1995) recommends the use of default Type M in the absence of specific information. A more detailed review is being carried out for a forthcoming ICRP Document on Occupational Intakes of Radionuclides (OIR), which will be issued following publication of the new Recommendations of ICRP. The then current (2006) draft of the OIR Document also recommended the use of default Type M for workers in the absence of specific information. The assumption of Type M was therefore adopted in all dose assessments here.

Recently, Harrison et al (2007) analysed the results of measurements (urine excretion and post-mortem tissues) of a case of accidental inhalation of ^{210}Po (form not stated) by a worker, reported by Ilyin (2001). A consistent fit to the data was obtained by using Type M default parameter values, except for the rapid fraction, f_r, for which a value of 0.4 was chosen instead of the default value of 0.1 (and also an f_1 value of 0.2 instead of the default value of 0.1, see 4.4.3.2 below). These parameter values are still consistent with assignment to Type M.

4.4.3.2 Absorption from the GI tract

The f_1 value for workers (0.1) was chosen, as appropriate for simple chemical forms of polonium. This is used in ICRP Publication 68 for both ingestion and inhalation intakes by workers. (The latter refers to material cleared from the respiratory tract to the GI tract.) Higher values, 0.5 for adults, are used in ICRP Publication 67 for ingestion by members of the public, but based on consideration of polonium biologically incorporated in food. A value for f_1 of 0.1 is used in ICRP Publication 71 for inhalation of Type M (or F) forms of polonium by members of the public. Moreover, the dose assessed from a urine sample is not particularly sensitive to the f_1 value.

4.4.4 Aerosol size

It was considered that ^{210}Po could possibly become airborne by mechanisms which might include:

- Suspension, resuspension, or evaporation of the original source material used in the poisoning.
- Following serious internal contamination, resuspension from drops of sweat and other activity on surfaces, including clothes, suspension in dust (such as flakes of skin), evaporation of sweat and exhalation as vapour. For health care workers, family and friends, there was also the possibility of aerosolisation of contaminated body fluids, for example during vomiting or urination, episodes of diarrhoea.

It was initially considered that airborne ^{210}Po originating from Mr Litvinenko might be of molecular size and so attach to the ambient room aerosol, as do radon decay products (e.g. ^{218}Po). The particle size distribution could therefore be that of the ambient room aerosol. Reference values for such aerosols were taken from Marsh *et al* (2002) and relate to a tri-modal aerosol (Table 4). This size distribution was initially assumed in special assessments for groups of people who could have been in Mr Litvinenko's presence, e.g. his family and friends, and health care workers. Assumption of this ambient aerosol size distribution gives an approximately three-fold higher dose assessed from a urine sample taken within a few weeks of intake than assumption of a 5-μm AMAD aerosol (Figure 10). In some assessments carried out later, the ICRP default assumption of a unimodal log-normal aerosol was used, either:

- 5-μm AMAD, the default assumption for occupational exposure, which is representative of exposures close to the source of an aerosol produced by dispersion processes (e.g. resuspension of dust, abrasion) or;
- 1-μm AMAD, the default assumption for environmental exposure of the public, and is representative of exposures remote from a source.

Appropriate deposition fractions were calculated for males and females, but the differences are small and have relatively little effect on the dose assessed from a urine sample.

Table 4 Size distribution of ambient room aerosol (Marsh et al 2002)

Fraction by activity	AMAD μm	Geometric standard deviation	Hygroscopic growth factor (applied to AMAD)	Density g cm^{-3}	Shape factor
15%	0.05	2.0	1.75	1.07	1.0
83%	0.23	2.1	2.0	1.05	1.0
2%	2.5	1.5	1.25	1.2	1.2

4.4.5 Time course of intake

The assumed time course of intake was derived from knowledge of the movements of the individual who provided the sample, combined with assumptions about the likely source of contamination. In cases where the time of intake was only known to lie between times t_1 and t_2, e.g., medical staff caring for Mr Litvinenko, the assumption of a constant chronic intake was made (Puncher et al 2006). Where information indicated that intake occurred at a specific time (e.g. a visit to a restaurant), the assumption of an

acute intake was made. The assumed time of intake has a relatively small effect on the assessed dose (see Figure 10).

4.4.6 Route of intake: inhalation versus ingestion

4.4.6.1 Sensitivity of assessed dose to assumed route of intake

The assumption made about the route of intake could make a large difference to the assessed dose per unit activity measured in urine, as shown in Figures 10 and 12, and Table 5. For example, 1 Bq measured in a 24-hour sample taken 20 days after an acute intake, would give assessed committed effective doses of:

- 26 mSv (ambient tri-modal aerosol, as defined in Table 4 and used for initial special assessments);
- 9 mSv (5-µm aerosol, as used in standard Category 2 assessments, Section 4.3.2);
- 0.8 mSv (ingestion);

Table 5 Sensitivity of dose per unit measurement to model assumptions

				Systemic model	
Aerosol size	Absorption Type	Inhalation (%)	Ingestion (%)	ICRP Public-ation 67	Leggett and Eckerman, 2001
				mSv*	mSv*
5 µm AMAD	F	100	0	0.8	1.1
5 µm AMAD	M	100	0	9.1	9.2
1 µm AMAD	M	100	0	16	13
Ambient**	M	100	0	26	17
Ambient	M	67	33	12	11
Ambient	M	50	50	7.7	8.2
Ambient	M	33	67	4.7	5.7
		0	100	0.8	1.4

* Dose (mSv) assessed from 1 Bq d^{-1}excreted in urine at 20 days after intake

** Ambient: Trimodal distribution typical of room air as defined in Table 4

Table 5 also gives corresponding doses assessed using the Leggett and Eckerman (2001) systemic model. In this model, there is greater urinary excretion following intake by inhalation (or wound) than following ingestion, provided by the "Plasma 2" compartment in Figure 2. Compared to use of the ICRP Publication 67 model, the assessed dose is higher following ingestion, but can be lower following inhalation. For a mixture there is little difference.

Table 6 shows how the dose conversion factor (dose per unit measurement) for most sets of assumptions listed in Table 5 (for the ICRP Publication 67 systemic model), changes with time between intake and sampling, for times up to 200 days. The 5-µm AMAD Type F aerosol is not shown because the results are very similar to those for ingestion. The values for 5-µm and 1-µm AMAD, 50% 1-µm AMAD + 50% ingestion, 100% ambient, and 100% ingestion, correspond to curves in Figure 10, but for simplicity of presentation values are given in mSv per *Bq* d^{-1} in Table 6 rather than mSv per *100 mBq* d^{-1} as in Figure 10.

The values at 20 days in Table 6 correspond to those in Table 5 for the ICRP Publication 67 systemic model.

The column for 5-μm AMAD in Table 6 also corresponds to the K-factor given in Table3, but in Table 6 the values are given in mSv per *Bq* d^{-1}, rather than mSv per *mBq* d^{-1}.

As noted above (Section 4.1, Figure 10), the dose assessed from a given measured value of activity in urine increases as the time between intake and measurement increases (Table 6). However, the increase is slower for inhalation than for ingestion. As a result, as time passes, the assessed dose becomes less sensitive to assumptions about the route of intake. Similarly, for the inhalation route, it becomes less sensitive to the assumed aerosol size.

Table 7 shows the intake assessed from a measurement of 1 Bq d^{-1} corresponding to each value in Table 6. The values for 5-μm and 1-μm AMAD, 50% 1-μm AMAD + 50% ingestion, 100% ambient, and 100% ingestion, correspond to curves in Figure 9, but for simplicity of presentation values are given in kBq per *Bq* d^{-1} in Table 7 rather than kBq per *100 mBq* d^{-1} as in Figure 9. As noted above (Section 4.1.2, Figure 9) the intake is remarkably insensitive to the assumptions about the route of intake.

4.4.6.2 Assumed route of intake

It was difficult to judge the likely ratio of intake by ingestion and inhalation, because of limitations in information available about the situation at the time of potential exposure.

It is generally assumed that for occupational exposure, inhalation is the most likely route of intake (e.g. ICRP, 1988), because working practices, especially in workplaces where radioactive materials are used, should minimise inadvertent ingestion. Typically, eating, drinking and smoking would not be permitted. Generally the assumption of inhalation leads to higher doses assessed from measurements on excreta than assumption of ingestion (Figure 10). The recently developed IDEAS Guidelines for assessing internal doses to workers from individual monitoring data suggest assuming 100% inhalation unless evidence for another route (ingestion or uptake through skin or wound) is available (Doerfel et al, 2006). For some potential exposure situations, the information available suggested that inhalation was likely to be the main route of intake. For example, personal protective measures and hospital hygiene should have minimised ingestion from hand-to-mouth contact for health care workers. For some hotel staff, dust was likely to be resuspended through activities such as sweeping and making beds.

However, the assumption of inhalation as the only route of intake is likely to lead to overestimates of dose in other potential exposure situations, especially those in bars and restaurants. Intake by mouth might have arisen there from hand-to-mouth contact after touching a contaminated surface or directly through contact with contaminated tableware. The aim was to make realistic assessments of dose, rather than overestimates. In view of the large difference in resulting dose, and uncertainty in the actual route, a mixture of inhalation and ingestion was assumed in most cases. The choice of a combination of these two routes of intake also reduced the chance that the 'true' dose was greatly under-estimated or over-estimated.

Table 6 Conversion factors, K, (mSv per Bq d^{-1}) for Category 3 assessments of dose from urine excretion rate, (see text for details)

Aerosol	5 µm	1 µm	1 µm	Ambient*	Ambient*	Ambient*	Ambient*	N/A
Inhalation,%	**100**	**100**	**50**	**100**	**67**	**50**	**33**	**0**
Ingestion, %	**0**	**0**	**50**	**0**	**33**	**50**	**67**	**100**
e(50) µSv Bq^{-1}	**2.2**	**3.0**	**1.6**	**3.3**	**2.2**	**1.7**	**1.2**	**0.25**
Day**								
5	7.3	14	5.0	26	10	6.3	3.7	0.6
10	7.9	15	5.4	26	10	6.7	4.0	0.6
15	8.4	15	5.8	26	11	7.2	4.3	0.7
20	9.1	16	6.2	26	12	7.7	4.7	0.8
25	10	17	6.7	26	12	8.3	5.1	0.8
30	10	17	7.2	26	13	8.9	5.5	0.9
35	11	18	7.8	27	14	10	6.0	1.0
40	12	19	8.4	28	15	10	6.5	1.1
45	13	20	9.1	29	16	11	7.0	1.2
50	14	21	9.8	30	17	12	7.6	1.3
55	15	22	11	31	18	13	8.3	1.5
60	16	24	11	32	19	14	9.0	1.6
65	18	25	12	33	20	15	10	1.8
70	19	26	13	35	21	16	11	2.0
75	20	28	14	36	23	17	11	2.1
80	22	29	15	38	24	18	12	2.4
85	24	31	17	39	26	20	13	2.6
90	25	33	18	41	28	21	14	2.9
95	27	35	19	43	29	23	16	3.1
100	30	37	21	46	31	24	17	3.4
105	32	39	22	48	33	26	18	3.8
110	34	41	24	50	36	28	20	4.2
115	37	44	26	53	38	30	21	4.6
120	40	46	28	56	40	32	23	5.0
125	43	49	30	59	43	35	25	5.5
130	46	52	32	62	46	37	27	6.1
135	49	55	35	65	49	40	29	6.7
140	53	59	38	68	52	43	32	7.3
145	57	62	40	72	56	46	34	8.1
150	61	66	43	76	59	49	37	8.8
155	66	70	47	80	63	52	40	10
160	70	74	50	85	67	56	43	11
165	76	78	54	89	71	60	46	12
170	81	83	58	94	76	64	50	13
175	87	88	62	99	81	69	54	14
180	93	93	66	105	86	74	58	16
185	100	99	71	111	92	79	62	17
190	107	105	76	117	97	84	67	19
195	115	111	82	123	104	90	72	21
200	123	118	88	130	110	96	78	23

* Ambient: Trimodal distribution typical of room air as defined in Table 4

**After presumed intake.

Table 7 Intake (kBq) assessed from excretion rate of ^{210}Po in urine (Bq d^{-1}) for assumptions used in Category 3 assessments

Aerosol	5 µm	1 µm	1 µm	Ambient*	Ambient*	Ambient*	Ambient*	N/A
Inhalation,%	**100**	**100**	**50**	**100**	**67**	**50**	**33**	**0**
Ingestion, %	**0**	**0**	**50**	**0**	**33**	**50**	**67**	**100**
Day**								
5	3.4	4.9	3.2	8.1	4.4	3.6	3.1	2.3
10	3.6	5.0	3.4	7.9	4.7	3.9	3.3	2.6
15	3.9	5.2	3.7	7.9	4.9	4.2	3.6	2.8
20	4.2	5.4	3.9	7.9	5.2	4.5	3.9	3.1
25	4.5	5.6	4.2	8.0	5.5	4.8	4.2	3.4
30	4.9	5.9	4.6	8.1	5.8	5.1	4.6	3.8
35	5.2	6.2	4.9	8.3	6.2	5.5	5.0	4.1
40	5.6	6.5	5.3	8.5	6.6	5.9	5.4	4.5
45	6.0	6.8	5.7	8.7	7.0	6.3	5.8	5.0
50	6.5	7.1	6.2	9.0	7.4	6.8	6.3	5.5
55	7.0	7.5	6.7	9.4	7.9	7.3	6.8	6.0
60	7.6	7.9	7.2	9.7	8.4	7.9	7.4	6.6
65	8.1	8.4	7.8	10	9.0	8.5	8.0	7.3
70	8.8	8.8	8.4	11	9.5	9.1	8.7	8.0
75	9.4	9.3	9.0	11	10	9.8	9.4	8.8
80	10	9.9	9.7	12	11	11	10	9.6
85	11	10	11	12	12	11	11	11
90	12	11	11	13	12	12	12	12
95	13	12	12	13	13	13	13	13
100	14	12	13	14	14	14	14	14
105	15	13	14	15	15	15	15	15
110	16	14	15	15	16	16	16	17
115	17	15	16	16	17	17	18	19
120	18	16	18	17	18	19	19	21
125	20	17	19	18	19	20	21	23
130	21	18	21	19	20	21	22	25
135	23	19	22	20	22	23	24	27
140	25	20	24	21	23	25	26	30
145	26	21	26	22	25	26	28	33
150	28	22	27	23	26	28	31	36
155	30	23	30	25	28	30	33	40
160	33	25	32	26	30	32	36	44
165	35	26	34	27	32	35	38	48
170	38	28	37	29	34	37	41	53
175	40	30	39	30	36	40	44	58
180	43	31	42	32	38	43	48	64
185	46	33	45	34	41	46	52	70
190	50	35	48	36	43	49	56	77
195	53	37	52	38	46	52	60	84
200	57	40	56	40	49	56	64	93

* Ambient: Trimodal distribution typical of room air as defined in Table 4.

**After presumed intake.

The assumptions were divided into categories, depending on the judgements made for different groups of individuals (Table 8). As noted above, it was often difficult in practice to decide which assumption to use in order to give a realistic assessment. In such cases, the cautious assumption of 67% inhalation and 33% ingestion was usually made. Note that the resulting factor (12 mSv from 1 Bq d^{-1} excreted in urine at 20 days after intake) is close to the arithmetic mean of those from assuming 100% inhalation and 100% ingestion.

Table 8 Assumed routes of intake and dose conversion factors for Category 3 assessments of dose from urine concentration

Route of intake	Assumption	Examples	Dose (mSv) assessed from 1 Bq d^{-1} excreted in urine*
Very likely to be inhalation	100% inhalation	Nurses trained in hospital hygiene caring for Mr Litvinenko	26
More likely to be inhalation than ingestion	67% inhalation & 33% ingestion	Chambermaids making beds in contaminated rooms; room cleaners; people close to Mr Litvinenko after he was contaminated (e.g. in a bar or restaurant at same time as Mr Litvinenko)	12
No specific information	50% inhalation & 50% ingestion	Hotel guests; room service waiters; workers in contaminated offices	8
More likely to be ingestion than inhalation	33% inhalation & 67% ingestion	People working in or visiting a bar or restaurant; but not on the day that Mr Litvinenko visited	5

*at 20 days after acute intake, assuming inhalation of ambient aerosol.

Following a special assessment, the calculated dose was reported. In some cases the dose was higher than that calculated by the standard assessment in the initial screening procedure, e.g., if 100% inhalation was assumed, because of the different size distribution. More usually the dose was lower than that calculated by the standard assessment. In some cases the assessed dose was less than 1 mSv, but nevertheless the calculated value was reported.

4.4.7 Current and future perspectives

At the time that this dose assessment protocol was developed, and when many of the assessments were carried out, available information was sparse. More information has since been obtained on possible events surrounding the incident, the distribution of surface activity in buildings, and the levels of internal contamination in individuals associated with those locations. Based on currently available information, some of the assumptions made may have overestimated intake by inhalation relative to ingestion. However, the assumption of inhalation rather than ingestion in the absence of information is consistent with practice in assessing occupational exposures and means that doses are more likely to be over-estimates than under-estimates. Furthermore, although uncertainties related to the route of intake have been highlighted here, there are other recognised sources of uncertainty in the assessment of doses from urine samples, for example the ratio of urinary to faecal excretion, and the biological retention time of systemic polonium. Since the risks estimated from the doses assessed from urine samples measured in this programme are all relatively small, the Health Protection Agency does not anticipate carrying out any general reassessment of doses. If the

exposures had resulted from a lawful practice involving the use of radioactive material, then further assessment would be appropriate as part of the investigation into whether the safety measures in place were adequate, but that is not the situation here.

5 RESULTS OF THE UK MONITORING PROGRAMME

Only a summary of results can be given here, because individual results are confidential to the people that provided the samples. It is however possible to provide some information on the distribution of results. Figure 13 summarises the results obtained in the public health programme for UK residents. Of the 753 measurements, 139 were below the minimum detectable activity (MDA, Section 2.2). Thus in those samples ^{210}Po was not positively identified, and the result is effectively an upper limit on the amount of ^{210}Po present. In most cases this was less than 10 mBq d^{-1}, and only one was greater than 30 mBq d^{-1} (because of the small sample volume it could only be reported as <100 mBq d^{-1}). For the remaining 614 measurements in which ^{210}Po was measured, there were 474 below the Reporting Level (RL) of 30 mBq d^{-1}, and most of these (416) were below 20 mBq d^{-1}, consistent with the expected range of natural background (Section 2.1). Nevertheless, 139 were above the RL, showing the likely presence of some ^{210}Po from the incident. Of these, 92, 41 and 6 were in the ranges 30–100, 100–1000 and >1000 mBq d^{-1}, respectively.

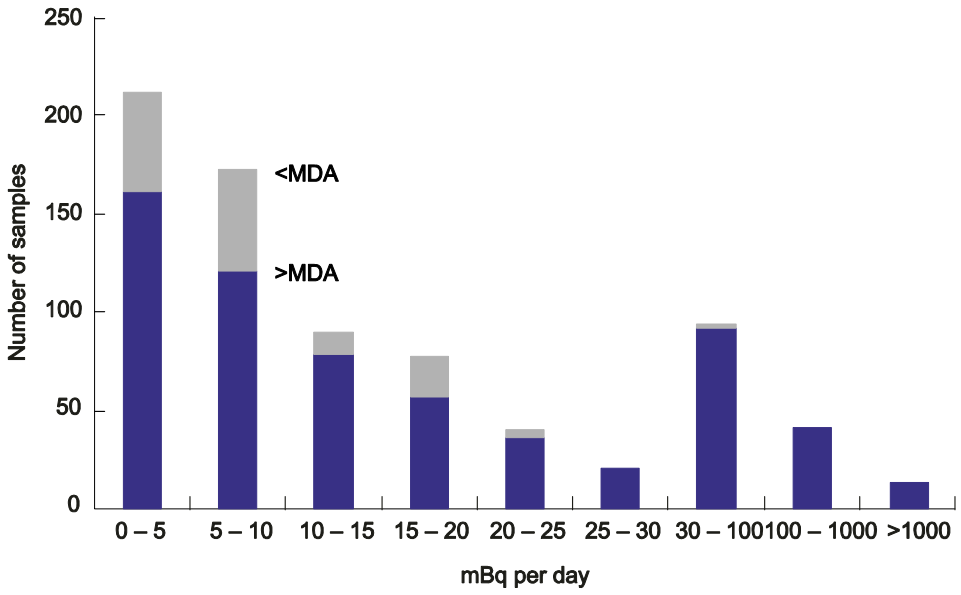

Figure 13 Activity distribution for all samples obtained in the public health programme for UK residents. The solid and open bars show results above and below the minimum detectable activity (MDA, Section 2.2), respectively. Thus the column labelled 5 – 10 mBq per day represents 121 results in the range 5 – 10 mBq per day and 52 results below a MDA in the range 5 – 10 mBq per day. Results in the latter group could actually have been less than 5 mBq per day.

In addition to the measurements made in this public health programme, urine monitoring was carried out on responders (e.g., police and those involved in environmental monitoring) by their employers. Results of this occupational health monitoring are not generally reported here. However, 24 measurements were made on urine samples from 17 members of Health Protection Agency staff (seven gave samples on two separate occasions). Three were control samples measured in the process of validating the measurement procedure (Section 2.2). The rest followed involvement in the environmental monitoring programme. Of these, 23 results were below the Reporting Level (RL), and one (4%) was Category 2 (< 1 mSv), supporting the working assumption, based on reports in the literature (Section 2.1), that excretion rates in the UK population are typically less than 30 mBq d^{-1}.

Table 9 gives a summary of results according to the type of location and occupation of the people who provided the samples. Some general features may be noted from the results in Table 9 (only groups with at least 15 individuals are considered):

- Likely contamination from the incident (results above the RL) was found in people potentially exposed in many locations.
- All the results for the restaurant customers, a group of more than 20 people, were below the RL, again supporting the working assumption that measurements above the RL are unusual in UK residents and indicate likely contamination from the incident.
- The proportion of results above the RL was relatively high (>35%) in four groups: Mr Litvinenko's family and friends; office staff; guests at Hotel A; and hotel bar staff. In other groups it was broadly similar (about 5 – 25%).
- The proportion of Category 3 results was also relatively high (>15%) in the same four groups. In most of the others it was broadly similar (about 3 – 10%), but it was relatively low in health care workers (1%).
- Many of the Category 3 results (26 out of 53) were associated with potential exposures in a hotel bar (staff and customers), including 11 out of the 17 in Category 3b.

Table 9 Results of measurements of 24-hour ^{210}Po urinary excretion and assessed doses, by group of exposed people (locations and occupations): numbers of measurements (n) in each Category (Cat)

	Number of measurements (n)							% of total	
Group	Total samples	<RL (Cat 1)	≥ RL and <1 mSv (Cat 2)	≥ 1 mSv and <6 mSv (Cat 3a)	≥ 6 mSv (Cat 3b)	≥ RL (Cat 2&3)	≥ 1 mSv (Cat 3)	≥ RL (Cat 2&3)	≥ 1 mSv (Cat 3)
Hospital A	36	32	3	1	0	4	1	11	3
Hospital B	40	35	5	0	0	5	0	13	0
Ambulance staff	2	1	1	0	0	1	0	50	0
Healthcare total	**78**	**68**	**9**	**1**	**0**	**10**	**1**	**13**	**1**
Family, friends, visitors	19	9	7	2	1	10	3	53	16
Offices									
– staff	15	9	2	4	0	6	4	40	27
– visitors	33	31	1	1	0	2	1	6	3
Restaurant A									
– staff	31	28	3	0	0	3	0	10	0
– customers	22	22	0	0	0	0	0	0	0
Restaurant B									
– staff	19	15	2	2	0	4	2	21	11
Hotel A									
– staff	110	88	15	4	3	22	7	20	6
– guests	19	12	4	3	0	7	3	37	16
Hotel B									
– staff	86	70	13	1	2	16	3	19	3
– guests	2	1	1	0	0	1	0	50	0
Hotel C									
– staff	21	16	3	2	0	5	2	24	10
– guests	25	23	1	1	0	2	1	8	4
Hotel bar									
– staff	16	3	4	4	5	13	9	81	56
– customers	256[a]	218	21	11	6	38	17	15	7
Non-healthcare total	**674**	**545**	**77**	**35**	**17**	**129**	**52**	**19**	**8**
Total	**752**	**613**	**86**	**36**	**17**	**139**	**53**	**18**	**7**

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 mBq d^{-1}.

6 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 set

up at the Health Protection Agency's Headquarters at Holborn Gate with up to seven staff from the Agency's Centre for Infections and LaRS. A summary of its work is given here: further details are being published elsewhere (Shaw et al in press). 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 also provided advice to other countries on sampling and dose assessments where requested. For example a document describing the dose assessment procedures used at the Health Protection Agency was sent to 10 overseas institutes in December 2006.

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 176 results obtained, 13 were greater than the Reporting Level (RL) of 30 mBq d^{-1}, eight giving assessed doses less than 1 mSv, and five in the range 1 to 6 mSv. Based on available information, individuals were placed in one of three categories of potential exposure risk: 'higher' (known to have visited a contaminated location and for whom urine testing would have been recommended if they had been UK residents), 'lower' (known not to have visited such locations) and 'unknown' (insufficient information to determine status). Nine of the 48 (19%) at 'higher' risk had results above the RL, a similar proportion to that in UK residents tested (see Section 5), most of whom would have been regarded as at 'higher' risk. For those at 'unknown' or 'lower' risk the fraction was only 3% (4/128), supporting the assumption that results above the RL indicate probable contact with ^{210}Po in this incident.

As noted in Section 3.3, when it became apparent that numbers of samples could exceed the RPD's analytical capacity, staff contacted other laboratories in the UK that carry out low-level measurements of ^{210}Po in environmental samples. As a back-up, several laboratories in Europe were also contacted and sent check samples to confirm consistency of results.

7 OBSERVATIONS AND CONCLUSIONS

Specialist knowledge of the behaviour and effects of incorporated radionuclides in general, and of ^{210}Po in particular, enabled a rapid assessment to be made of the amount of activity likely to have been involved in the poisoning of Mr Alexander Litvinenko, and the potential implications for public health.

It was rapidly established that individual monitoring through urine sampling had the capability to assess intakes of ^{210}Po and resulting doses well below those likely to cause observable health effects.

A system for monitoring hundreds of people per week was developed based on techniques used for low-level measurements of ^{210}Po in environmental samples. A monitoring programme producing reliable results was rapidly put into operation.

A massive effort is required in the first hours to days of such an incident in order simultaneously to give specialist advice, decide on procedures, set up systems for monitoring large numbers of people, collate and provide summaries of results, and process early samples rapidly to inform the response and give specialist advice. The Agency's response in this respect benefited from experience of the programme of nuclear emergency and counter terrorism exercises, and training of staff to fulfil a variety of roles.

The ability of the Agency to develop and undertake this programme of individual monitoring stemmed from well established laboratory and modelling capabilities. Such capabilities need to be maintained to enable an effective response to unpredictable radioactive contamination situations.

The scale of the necessary monitoring programme went beyond the capacity of the Agency's laboratories, or indeed any single laboratory in the UK. It was important to have had the rapid co-operation of other organisations with the capability and capacity to support the response. This co-operation benefited from the experience of working together in emergency exercises and other networking activities such as participation in quality assurance intercomparison exercises.

Although it proved not to be needed, there was the prospect of requesting the support of overseas laboratories to deal with the volume of monitoring. The positive attitude of those laboratories that were contacted, as a contingency, was appreciated and demonstrated the value of international network arrangements.

There is a need for a pre-prepared, but flexible database to enable information on the same sample to be entered at different locations.

The large number of measurements and the need for rapid reporting of results provided a major challenge. To address this, a categorisation 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. This proved to be successful.

Pragmatic decisions had to be made, in a short period of time, on the criteria underpinning the dose categories. However it needs to be emphasised that other criteria could have been justified and that in any future radiation incident different criteria may be appropriate, depending on the specific circumstances such as the number of people being measured.

There is a need for material prepared in advance to facilitate providing people with their results, and which also explains the significance of the results of monitoring in terms of risk.

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