ABSTRACT

On November 1, 2006, former Russian agent Alexander Litvinenko suddenly fell ill and was hospitalised. He died three weeks later, becoming the first known victim of lethal polonium-210 induced acute radiation syndrome. Po-210 is an alpha emitter and is not a radiological hazard as long as it remains outside the body. If taken into the body, much of Po-210 is subsequently excreted, mostly through faeces and some through urine and other pathways. After uptake by the blood, Po-210 is widely distributed through soft body tissues including bone marrow. The internal dose from polonium in the body gives rise to an increase in lifetime cancer risk. Very high radiation doses can cause severe damage to body tissues and organs and in the extreme can be fatal. The hazard function model was used to estimate the lethal levels of intake of Po-210. In case of Litvinenko the destruction of bone marrow and resulting failure of the immune system was most probably the main cause of death, likely to be compounded by damage caused by higher doses to other organs, including kidneys and liver. It was estimated that the ingestion of several hundreds MBq or just about a microgram of Po-210 can be lethal.

1 INTRODUCTION

On November 1, 2006, former Russian agent Alexander Litvinenko suddenly fell ill and was hospitalised. He died three weeks later, becoming the first known victim of lethal polonium-210 induced acute radiation syndrome.

The timeline of events was presented in [1]. On November 1, 2006, Litvinenko had two meetings and one of them is suspected to have been fatal to him. In a hotel bar in London he was poisoned by Po-210 by drinking the contaminated tea. On the same day vomiting, as one of the first symptoms of acute radiation syndrome appeared and was followed the next day by diarrhea. Three days after the poisoning he was hospitalised. In the first period he was treated symptomatically because his illness and possible causes were not diagnosed. After 14 days, when his hair started to fall out, the possibility of thallium poisoning was considered. The laboratory tests showed only insignificant thallium levels in his body. The suspicion aroused that maybe his illness was caused by radioactive thallium. The measurements of the radioactivity in his body were negative, but they were performed by standard radiation detectors, which can only detect penetrating gamma radiation. Only a few hours before his death special laboratory tests revealed the presence of Po-210 in his body.
The Health Protection Agency (HPA) immediately started to search for places, potentially possible to be contaminated with polonium. Most of them were the locations that Litvinenko visited after his poisoning. Some of them could be related to the presence of the men who presumably poisoned him, or to the presence of people that had been in close contact with Litvinenko. Additionally HPA started to monitor people who had been in close contact with Litvinenko or had been present at contaminated places. In some of these cases Po-210 was found in their bodies but in no case its activity was significant enough to present any danger for an acute radiation syndrome. Even the lifetime risk for a cancer in these people was estimated to be very low.

In this presentation the basic information about polonium is given, its internal dosimetry is discussed from the point of view of risks for induction of cancer and as the cause of acute radiation syndrome. The hazard function model for risk of deterministic effects is presented and used for the assessment of ingested activity of Po-210 that may be lethal.

2 PROPERTIES, PRODUCTION AND USE OF POLONIUM-210

In 1896 Henri Becquerel discovered natural radiation in form of the so called "uranium rays", the penetrating radiation emitted by uranium salts and detected by use of photographic plates. The word "radioactive" appeared for the first time in 1898 when Marie and Pierre Curie announced the discovery of polonium-210, separated from the pitchblende ore that contained uranium.

More than 30 isotopes of polonium are known with atomic masses ranging from 188 to 230. All polonium isotopes are radioactive, but most are very short lived. Only three polonium isotopes have relatively long half-live: Po-208, Po-209 and Po-210. Their main properties are shown in table 1. Polonium-208 and Po-209 are produced artificially and are used as tracers in radiochemical determination of Po-210.

Po-210 occurs naturally in uranium ore, being one of the daughters in the U-238 decay series. One ton of uranium ore contains only about 100 micrograms of polonium. It is also found in very low concentrations in the earth's crust and there are tiny amounts in air and our bodies. Due to its scarcity, polonium is usually produced by bombarding a stable isotope bismuth-209 with neutrons in a nuclear reactor. This forms bismuth-210, which has a half-life of 5 days. Bismuth 210 decays into Po-210 through beta decay. Only about 100 grams are produced worldwide each year, practically all of it in Russia, making polonium exceedingly rare.

Table 1: Properties of long-lived polonium isotopes

<table>
<thead>
<tr>
<th>Isotope</th>
<th>t_{1/2}</th>
<th>E_α (MeV)</th>
<th>E_γ (keV)</th>
<th>b_γ</th>
</tr>
</thead>
<tbody>
<tr>
<td>Po-208</td>
<td>2.90 a</td>
<td>5.1</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Po-209</td>
<td>102 a</td>
<td>4.9</td>
<td>896</td>
<td>2.5 E-5</td>
</tr>
<tr>
<td>Po-210</td>
<td>138 d</td>
<td>5.3</td>
<td>803</td>
<td>1.1 E-5</td>
</tr>
</tbody>
</table>

Po-210 is essentially an alpha emitter that has a half-life of 138 days. By emitting an alpha particle with the energy of 5.3 MeV it decays in the stable daughter isotope Pb-206. About one in 100,000 decays results in the emission of a gamma ray with the energy of 803 keV. Therefore it is impossible to detect small, although radiologically significant quantities of Po-210 by standard radiation monitors that are, for instance, used for control of illicit trafficking with radioactive materials. For the same reason, if properly sealed, it does not pose any radiological hazard to the person carrying it. Po-210 has a specific activity of 166 TBq per gram that is 4500 times as much as in Ra-226. The energy released by its decay is so large...
(140 W·g$^{-1}$) that a capsule containing half a gram reaches a temperature above 500°C. Po-210 has been used as a lightweight heat source to power thermoelectric cells in satellites.

Polonium mixed with beryllium was used in first atomic bombs as the neutron source that initiated the chain reaction (the so-called initiator). It had been also used as a neutron source in other applications, but its relatively short half-life proved to be a serious drawback in these cases.

Po-210 is still used in some industrial applications such as static eliminators, which are devices designed to eliminate static electricity in processes such as rolling paper and manufacturing sheet plastics.

Because it emits alpha particles, Po-210 represents a radiation hazard only if it gets into the body by inhalation, ingestion or through a wound.

3 DOSIMETRY OF POLONIUM

Po-210 is essentially a pure alpha emitter and it is not a radiological hazard as long as it remains outside the body. If it gets into the body, Po-210 is subsequently excreted, mostly through faeces but some is excreted through urine and other pathways. After uptake to blood, Po-210 is widely distributed though soft body tissues. The biokinetic model for polonium, as proposed by ICRP [2], is shown in Figure 1. If taken by ingestion, 90 % of the activity is excreted by faeces and urine and the remaining 10 % is concentrated in liver (30 %), red bone marrow (10%), kidneys (10%), spleen (5%), and in other tissues (45%). Thus ingestion of polonium causes rather uniform whole-body exposure. The biological half-life in body tissues is approximately 50 days and the resulting effective half-life is 37 days.

Radiation doses are assumed to give rise to an increase in lifetime cancer risk (stochastic effects of radiation). The larger the dose, the larger the risk. Very high radiation doses can cause severe damage to body tissues and organs and in the extreme can be fatal (deterministic effects of radiation).

Figure 1: Biokinetic behaviour of Po-210 (ICRP67)
3.1 Stochastic Effects of Po-210

The probability for stochastic effects is proportional to the effective dose $E$ or, in case of internal exposure, to the committed effective dose, which is based on the lifetime dose due to the incorporated radionuclide. Both quantities take into account the type and energy of radiation (radiation weighting factor $w_R$) and the susceptibility of organs and tissues for induction of cancers (tissue weighting factor $w_T$):

$$E = \sum_T w_T \cdot \sum_R w_R \cdot D_{T,R}$$

$D_{T,R}$ is the average absorbed dose (measured in $\text{Gy} = \text{J kg}^{-1}$) caused by radiation $R$ in the organ or tissue $T$. The unit for effective dose is $\text{J kg}^{-1}$, termed the sievert ($\text{Sv}$).

For alpha particles the radiation weighting factor equals to 20. In the case of Po-210 ingestion the committed effective dose is assessed by the conversion factor $2.4 \cdot 10^{-7} \text{ Sv Bq}^{-1}$. Thus the ingestion of 4 kBq or about 20 pg of Po-210 causes the committed effective dose of 1 mSv which is the annual limit for members of the public. The risk of a fatal cancer is typically taken to be about 0.005% per milisievert. This means that if 20,000 people each received an additional effective dose of one milisievert a single radiation-induced cancer death would be expected to occur in this group of the population.

In the past the intake of Po-210 was the most probable cause of death of Irène Joliot-Curie. In 1946 she had been accidentally exposed to polonium when a sealed capsule of the element exploded on her laboratory bench. She died of leukaemia ten years later.

In the case of Litvinenko's poisoning an extreme effort of the British authorities was also devoted to estimate the possible intake of polonium to access the radiation exposures and the resulting cancer risks to the people that had been in close contact with Litvinenko or happened to be at the contaminated places. An extensive screening of these persons had been conducted by Health Protection Agency [3]. From the end of November 2006 till March 15, 2007, 733 persons had been monitored by urine analysis and it was found out that all of them were exposed to the levels of Po-210 that cannot be of health concern or are, even in extreme cases, so low that any increased risk of a cancer in the long term is likely to be very small. In no case any acute risk of radiation was expected.

3.2 Deterministic effects of Po-210

In case of larger intake of Po-210, what occurred in Litvinenko's poisoning, a massive damage of several organs, where the isotope concentrates can be the cause of death. For Po-210 the destruction of bone marrow and resulting failure of the immune system is most probably the main cause of death, likely to be compounded by damage caused by higher doses to other organs, including kidneys and liver.

Risk of death $R$ for deterministic effects has been evaluated in [4] and [5] using the hazard function model:

$$R = 1 - e^{-H}$$

$H$ is the hazard function, depending on the absorbed dose in an organ or tissue, modified by the relative biological effectiveness of the radiation (RBE). It is called the adjusted dose (AD), measured in gray-equivalent (Gy-Eq). RBE replaces the radiation weighting factors used in radiation protection to estimate the risk of stochastic effects. The values of RBE are different and they are typically lower for deterministic effects. For alpha particles $\text{RBE}_\alpha$ has
been estimated to be 2 for bone marrow lethal damage. For internal exposure the hazard function depends on specifics of the target organ and the time of exposure.

\[
H = \ln(2) \left[ \int_0^{\ln(2) \tau} \frac{ADR_0}{AD_{50}} e^{-\frac{ln(2) t}{t_{ef}}} dt \right]^V
\]

where \(ADR_0\) is the initial adjusted dose rate, \(AD_{50}\) is the lethal adjusted dose for 50% of the population (estimated to be 3 Gy-Eq for bone marrow), \(V\) is the shape parameter of the sigmoid curve and defines its steepness (\(V = 6\) for bone marrow), \(t_{ef}\) is the effective half life of polonium in body tissues, and \(\tau\) is the time of exposure. For Po-210 ingestion \(ADR_0\) is given by expression (4):

\[
ADR_0 = \frac{E_{\alpha} \cdot RBE_{\alpha} \cdot f_1 \cdot f_{BM} \cdot A_{\text{ing}}}{m_{BM}} dt
\]

The risk for death from bone marrow syndrome for different ingested activities of Po-210 was calculated by use of equations (3) and (4). The following values for relevant parameters were used:

- \(E_{\alpha}\) energy of Po-210 alpha particles (\(E_{\alpha} = 5.3\) MeV)
- \(RBE_{\alpha}\) relative biological effectiveness of alpha particles for bone marrow lethal damage (\(RBE_{\alpha} = 2\))
- \(AD_{50}\) adjusted dose to bone marrow, which leads to 50% of deaths in a population (\(AD_{50} = 3\) Gy-Eq)
- \(V\) the shape parameter of the sigmoid curve (\(V = 6\))
- \(f_1\) the fraction of ingested activity that enters the bloodstream (\(f_1 = 0.1\))
- \(f_{BM}\) the fraction of the activity in the bloodstream that is taken-up by bone marrow (\(f_{BM} = 0.1\))
- \(m_{BM}\) the mass of the bone marrow in a reference man (\(m_{BM} = 1.5\) kg)
- \(t_{ef}\) effective half-life of ingested polonium (\(t_{ef} = 37\) days)
- \(A_{\text{ing}}\) the ingested activity of Po-210, free variable in calculations.

The time of integration \(\tau\) in expression (3) was set to 22 days, which is the time that elapsed between the polonium poisoning of Litvinenko and his death. The dependence of the risk of death from bone marrow destruction on ingested activity of Po-210 is shown in Figure 2. About 50 MBq is the threshold activity and several hundred MBq is a lethal intake. In this case the ingested activity corresponds only to about a microgram of Po-210.

In case of Litvinenko the calculations based on the effective half-life of Po-210 show that only about 30% of final dose to the bone marrow had been delivered in 22 days. Therefore the ingested activity must have been supra lethal.
4 CONCLUSIONS

Polonium-210 is essentially a pure alpha emitter and presents a radiological hazard only if it gets into the body via ingestion, inhalation or through wounds. After intake by ingestion most of Po-210 is subsequently excreted but about 10% of the activity is accumulated in soft tissues like bone marrow, kidneys, liver, and spleen. Its biological half-life is 50 days, resulting in the effective half-life of 37 days.

The internal dose from polonium in the body gives rise to an increase in lifetime cancer risk. Very high radiation doses can cause severe damage to body tissues and organs and in the extreme can be fatal.

In case of Litvinenko's poisoning with Po-210 the destruction of bone marrow and resulting failure of the immune system was most probably the main cause of death, likely to be compounded by damage caused by higher doses to other organs, including kidneys and liver.

By use of the hazard function model it was estimated that the ingestion of only several hundreds MBq or just about a microgram of Po-210 can be lethal.

By all of its properties polonium-210 seems to be almost an ideal poison: tiny quantities ingested are sufficient to cause irreversible damage to the organism, it does not, if properly sealed, pose any radiological hazard to the carrier and is impossible to be detected by routine radiation monitoring. On the other hand the thorough investigation of English authorities, supported by the very demanding laboratory analyses could reveal the traces of Po-210 in suspected contaminated places and in persons who might have been exposed to polonium.
REFERENCES


[2] ICRP Publication 67: Age-Dependent Doses to Members of the Public from Intake of Radionuclides

