Am-241 as a Metabolic Tracer for Inhaled Pu Nitrate in External Chest Counting

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INTRODUCTION
The most difficult radionuclides to measure accurately by external chest counting are the isotopes of plutonium. They are detected through weak emission of low energy L X-rays; in the case of 239Pu, 11-20 keV L X-rays are emitted with the total emission rate of 0.042 per disintegration. These photons are severely attenuated by overlying tissue; typically about 0.1 % of the photons, that is, only 0.00004 photon per unit α-disintegration will escape from the human thorax.

The plutonium treated in nuclear fuel cycle is usually accompanied with 241Am produced from 241Pu by β disintegration, except for the plutonium freshly separated by chemical processes. The 241Am emits γ-rays of 60 keV with the emission rate of 0.36, which are more penetrable than the L X-rays. The 241Am could improve the detection limit of chest counting of Pu, if it being used as a metabolic tracer for Pu in lungs. Furthermore the error in the estimation of chest wall thickness dose not result in an error so large as for plutonium in the estimation of the deposited activity.

In the ICRP dosimetric model, americium is assigned to Type M for all chemical forms (1). On the other hand, plutonium is assigned to Type M or Type S, depending on its chemical forms (1). A key problem to use 241Am as a tracer of inhaled plutonium is thus whether the ratio of americium vs. plutonium changes or is kept constant in the lungs.

It has already been shown that the 241Am which would probably be embedded in a matrix of PuO2 is cleared from the lungs at the same rate as the Pu for long time after intake (2), which supports the validity of 241Am as a tracer of inhaled Pu in external chest counting. In the present work another possible chemical form present in workplaces, Pu nitrate, has been investigated.

EXPERIMENTAL
(a) Aerosol generation and inhalation
The facility of aerosol generation/inhalation was constructed in interconnected gloveboxes (3). The solution of Pu(NO3)4 was nebulized using a compressed-air operated nebulizer. The resultant droplets were passed through a tube with fresh air and conducted into a multi-port nose-only exposure chamber, which resulted in polydisperse aerosols of Pu(NO3)4 with 0.6 µm in AMAD. The size of aerosols was determined with a cascade impactor. The activity ratio of 241Am vs. Pu in the solution was 2.4 %.

Twenty rats can be exposed simultaneously to the aerosols without anesthesia.

(b) Experimental animals
Young adult male Wistar rats, being 13 weeks old and weighing 230 g at the time of exposure were used. Forty exposed rats were used for this purpose. They were divided into 8 groups.

(c) Radioactivity measurement
The exposed rats were periodically sacrificed and the radioactivity of 241Am and 238/239/240Pu in the autopsied lungs were measured. Photon spectrometry with a NaI/ CsI phoswich detector system was applied for the measurement of radioactivity of 241Am and 238/239/240Pu in the autopsied lungs.

The phoswich detector system was calibrated by two steps. At the first step, the system was calibrated for a point source as usual calibration procedures using a commercially available point source of 241Am and a self-made calibration point source of 238/239/240Pu. Next, the correction of counting efficiency was carried out by Monte-Carlo simulation for self-absorption of low energy photons and solid angle effects of volume source.

The minimum detectable radioactivity of the system for 238/239/240Pu is 0.6 Bq for the standard error of 33 %. The details of this counting system is given elsewhere (4).

Time course of lung retention of 238/239/240Pu for living rats was followed by in vivo counting.
A rat contained in a plastic box with being zipped in a polyethylene bag was placed in a shielded chamber with a 50 mm thick wall of lead with a lining of a 5 mm plates of copper and acrylic resin plate, being 1290mm(L) x 590mm(W) x 740mm(H) in outer size and 0.3 m$^3$ in capacity. Three thin NaI(Tl) scintillation detectors, 51 mm in diameter and 1mm in thickness were housed within this shielded chamber and were located closely to the ventral side and to both the lateral sides of the rat. The usual counting time was 1000 s. The minimum detectable activity was 30 Bq for 33% relative uncertainty for a rat of 230 g weight.

The details of the \textit{in vivo} counting system is given in the previous paper (5).

The schedule of the radioactivity measurement is shown in \textbf{Table 1}.

| Schedule of radioactivity measurement |
|-------------------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| 1G                            | 2G                | 3G                | 4G                | 5G                | 6G                | 7G                |
| 1st day                       | Autopsy           |                   |                   |                   |                   | In vivo |
| 2nd day                       | Autopsy           | In vivo           | In vivo           | In vivo           |                   |                   |
| 3rd day                       |                   |                   |                   |                   |                   |                   |
| 28th day                      |                   |                   |                   |                   |                   |                   |
| 84th day                      |                   |                   |                   |                   |                   |                   |
| 168th day                     |                   |                   |                   |                   |                   |                   |
| 252nd day                     |                   |                   |                   |                   |                   |                   |
| 336th day                     |                   |                   |                   |                   |                   |                   |
| Autopsy                       |                   |                   |                   |                   |                   |                   |

\textbf{RESULTS AND DISCUSSION}

(a) Photon spectrum from $^{241}$Am and $^{238/239/240}$Pu in an autopsied lung

\textbf{Figure 1} shows an energy spectrum of photons obtained for an autopsied lung. Two peaks are observed around 17 keV and 60 keV; the former resulted from L X-rays of $^{239}$Pu and $^{241}$Am and the latter the $\gamma$-rays of $^{241}$Am.

![Fig. 1: Photon spectrum from $^{241}$Am and $^{238/239/240}$Pu in an autopsied lung](image)

The detector energy resolution for the 60 keV $\gamma$-ray region was 12% and the apparent resolution for the L X-ray region was 42%. The background counts of this system were 0.09 cps for L X-ray region and for $\gamma$-ray region, too.
(b) Distribution of deposited radioactivity among different lung lobes

Figure 2 shows the distribution of initially deposited radioactivity of $^{238/239/240}$Pu among different lung lobes; Fig. 2 (a) is for the fraction of radioactivity deposited in each lung lobe and (b) for the activity concentration (radioactivity per unit mass) of each lung lobe relative to the mean activity concentration of the whole lung.

The fraction of radioactivity deposited in each lung lobe differed much between the lobes. On the other hand, the concentration of radioactivity was nearly the same in all the lobes. This experimental result supported the evenness of deposition probability within a whole lung.

The evenness of the activity concentration was maintained at least up to a half year after the exposure, which indicated that the clearance of Pu nitrate occurred at the same rate in every lobes.

(c) Retention function of inhaled Pu nitrate

The result of the follow-up of lung retention of $^{238/239/240}$Pu for half a year that was measured by in vivo
counting is shown in Fig.3. The solid lines are for the rats exposed at higher level of radioactivity (2400 Bq on an average) and the dotted lines are for the rats exposed at lower level of radioactivity (900 Bq on an average). The present result shows that the alveolar lung clearance of Pu nitrate was independent of initial lung burden.

(d) Ratio of $^{241}\text{Am}$/$^{238/239/240}\text{Pu}$ in lungs

The ratio of $^{241}\text{Am}$/$^{238/239/240}\text{Pu}$ was obtained from the measured lung content of each radionuclide. The result is shown in Fig.4.

The activity ratio of $^{241}\text{Am}$ vs. Pu in lungs, 2.4 % at the exposure, slowly decreased to 2.1 % at 4 weeks and 2.0 % at 24 weeks. However, from practical point of view, it could be concluded that $^{241}\text{Am}$ would be a valid metabolic tracer for inhaled Pu nitrate at least for half a year post inhalation, considering the detection limit for Pu in chest counting, which is much greater than the ALI of Pu and the large uncertainties due to the estimation of chest wall thickness, the change in the distribution of particles within the lungs etc.
CONCLUSION

As well as for Pu dioxides, the validity of $^{241}$Am as a metabolic tracer of inhaled Pu in external chest counting was supported from practical point of view by the experiment using rats exposed to the aerosols of Pu nitrates which contain 2.4 % $^{241}$Am in activity.

REFERENCES