TWENTY-YEAR FOLLOW-UP OF A Pu/Am INHALATION CASE

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In 1983 a technician inhaled a mixture of Pu/Am aerosols in an accidental situation in the hotlab of Paul Scherrer Institute (PSI). This case is of interest for long-term follow-up since the technician was relatively young (26 y) at the time of intake, no chelating agent was used to alter retention and excretion and the inhaled activity was rather high (≈ 20 kBq of alpha emitters). The results obtained from periodic lung counts, urinary and faecal excretions as well as from some bone and liver measurements up to the year 2003 are presented. The measurements were mainly made at PSI but also at FZK Karlsruhe, Germany, and PNNL Hanford, USA. The evaluation and dose estimation of this case was done by several institutions, such as FZK, PNNL and NRPB in addition to PSI. Elements of the case were used in international biokinetic model validation programs by EURADOS/EULEP and IAEA and the ^{241}Am data are given as example in Annex E of the ICRP 'Guide for the Practical Application of the ICRP Human Respiratory Tract Model'. An overview is given on the various results obtained by the different institutions using their models and methods for interpretation of the measured data. While estimation of intake varies by more than an order of magnitude, final estimation of effective committed dose varies only in the range of 0.5–1.5 Sv.

THE ACCIDENTAL SITUATION IN 1983

The former Swiss Federal Institute for Reactor Research (EIR), now Paul Scherrer Institute (PSI) operated a hot lab where research on nuclear fuel has been performed for more than a decade. As a consequence of the production of uranium/plutonium carbide microsphere fuel elements some liquid waste resulted. The volume of this liquid waste was reduced periodically by evaporation in a special apparatus within a glove box of the hot lab. On one occasion an incident happened as described in the original EIR report[(1)]:

> At 16:15 on 24 May 1983 a rapid chemical reaction occurred in an apparatus in which a routine waste sludge evaporation process was under way. The waste was produced during fuel fabrication. The resulting pressure wave ruptured a number of parts at the glove box in which the equipment was situated leading to an alpha contamination of that and neighboring laboratories. The alarm system activated by the fire alarm, the activity detectors and manually by the operators functioned correctly.

A total of seven persons working in the immediate surrounding of the place of the incident inhaled a detectable amount of alpha emitting radionuclides. For one person only the estimated committed dose was above the legal limits. This refers to a young man of 26 y at the time of the incident (M.D., born in 1957). All data on measurements and analysis are given for this person only.

The material dispersed at this incident was a mixture of different waste products in various chemical forms. Therefore, no detailed information could be given on the chemical form and the particle size distribution of the inhaled radioactive material. The major component of the fuel material used at that time was natural uranium and a plutonium mixture with the badge name P-13. The typical ratio by weight was ~80% of uranium and 20% of P-13. The activity of uranium was, therefore, several orders of magnitude lower than for plutonium. The material P-13 has a relative composition as given in Table 1. This composition was assumed to be applicable for the interpretation of the inhalation case.

Immediately after the incident the most affected person (M.D.) was transferred to the EIR decontamination facility. It turned out that his head and neck were heavily contaminated. A nasal swab and a bronchial slime probe were taken and after a first decontamination a chest count was performed at 19:00 h of May 24. These first measurements gave the following results: nasal swab, 5.5 kBq alpha activity; bronchial slime, 1.4 kBq alpha activity; chest count, 390 Bq ^{241}Am, corresponding to 3.9 kBq alpha activity (for P-13 material).

These results indicated the severity of the inhalation case and the collection of urine and faeces and a follow-up of chest counts was initiated. At that time a decision had also to be taken on the application of a chelating agent to enhance excretion. Ampoules with Na- and Ca-DTPA were available at the EIR decontamination facility for injection. It was assumed that the inhaled Pu was in an insoluble form and that diethylenetriaminepentacetate (DTPA) would not significantly reduce committed dose, even if it would enhance urinary excretion. Therefore, it was decided not to apply the chelating agents.

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THE MEASUREMENTS PERFORMED ON M.D.

In the 20-y follow-up of this case a total of >100 independent measurements were performed at five different laboratories (see Table 2). These measurements consisted of *in vivo* measurements of the chest, the lungs, the liver, the skeleton and the pulmonary lymph nodes, as well as urinary and faecal analysis. In 1992 a count of chromosomal aberrations in peripheral lymphocytes was also initiated. The results of these measurements are given in the Annexure.

In vivo measurements

The measurement techniques and the calibration methods for the *in vivo* measurements have changed dramatically since 1983. The first measurements were taken with phoswich NaI/CsI-detectors and later on intrinsic germanium detector arrays were used (planar n-type Ge-spectrometers). The three laboratories involved in this type of measurements (Lab 1, 2 and 3) did not directly compare their calibrations in a specific intercomparison exercise. However, all laboratories participated in international intercomparison programs using the Livermore phantom as a reference for chest and lungs. Some effort was made to determine the uncertainty especially of the very early measurements performed at Lab 1. Due to the fact that not all relevant information was completely stored, it was not possible to arrive at a standardised and uniform method for the expression of uncertainty for all these measurements. Therefore, it was decided to present the results without indication of uncertainties.

Table 1. Composition of the fuel material P-13 normalised to total alpha activity.

Radionuclide	Alpha activity	Beta activity
^{238}Pu	0.09	
^{239}Pu	0.55	
^{240}Pu	0.26	
^{241}Am	0.10	
^{241}Pu		7.50
Total	1.00	7.50

Urine and faecal analysis

Prior to the chemical separation procedures to preconcentrate trace levels of Pu and Am, procedures for sample dissolution are necessary. To destroy the organic matter in urine, Lab 1 added 100 ml 14 Mol/l HNO_3 to 1 litre samples. After covering with watch glass the solutions were then boiled for about 5 h until a clear solution was obtained. The faecal samples were initially converted to dry ash in a high temperature oven and the ash was then digested in a glass beaker using a mixture of HNO_3/H_2SO_4 under boiling and back-flow via use of a condensation tube. The clear solutions were then taken for radiochemical purification (next paragraph). For chemical yield determination ^{243}Am and ^{242}Pu tracers were added to the urine sample prior to oxidation and to the faecal samples after it was converted to dry ash.

Initially Lab 1 applied a gross alpha method for determination of transuranium isotopes via simultaneous adsorption of actinides on glass fibre materials. This method was later replaced by extraction chromatography methods using selective resins for consecutive separation of Pu and Am isotopes. The actinides of the urine samples are pre-concentrated onto a highly selective extractant commercially available as Actinide Resin (EiChrom SA, Darien, IL, USA) simply by adding 200 mg resin to a 0.5 litre sample and stirring for 1 h. The separation of the resin containing the actinides from the bulk of the solution is obtained via filtration on cellulose nitrate membrane filters. Stripping of the reagent from the inert support (polymeric substrate) is performed using organic solvents such as isopropanol. After decomposition on a heating plate the electrolytic

Table 2. Laboratories involved in measurements on the inhalation case of M.D.

Lab number	Name of laboratory	Type of measurements
Lab 1:	PSI (former EIR), Villigen/Würenlingen, Switzerland	Chest counts were performed until 1991 with two phoswich detectors installed in the PSI whole body counter. Urine and faecal analysis
Lab 2:	FZK (former KfK), Karlsruhe, Germany	Chest counts were performed with phoswich detectors and from 1991 lung, liver, bone and lymph node measurements were made with Ge detectors. Urine and faecal analysis
Lab 3:	PNNL (former PNL), Richland, USA	Lung, liver, skeleton and lymph node measurements were made with Ge detectors
Lab 4:	IT Corporation, Richland, USA	Urine analysis
Lab 5:	NRPB, Chilton, UK	Counting of chromosome aberrations

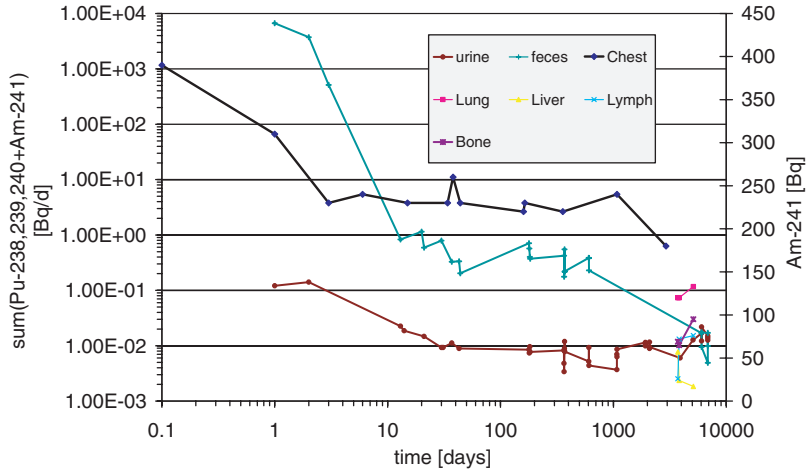

Figure 1. Results of *in vivo* (^{241}Am) and excretion (^{238,239,240}Pu and ^{241}Am) measurements in three different laboratories in the time period of 1983 until 2003.

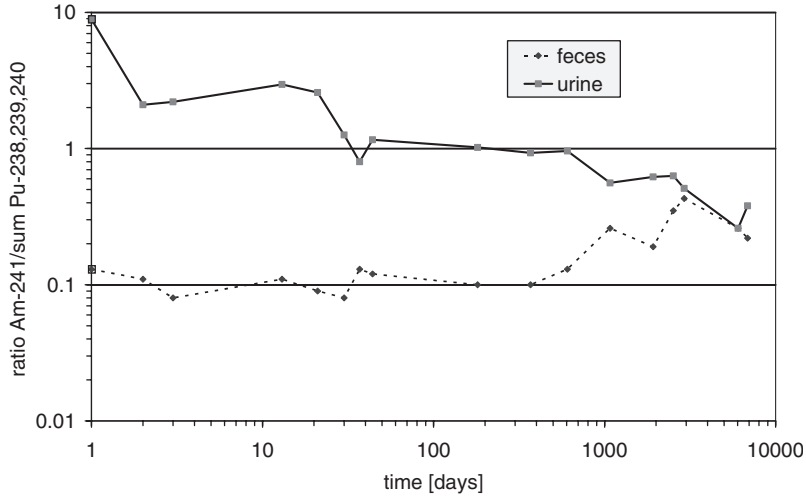

Figure 2. Ratio of americium to alpha emitting plutonium, in excretions of urine and faeces for a time period of 20 y after the inhalation.

deposition is carried out in a $NaHSO_4/H_2SO_4$ buffer solution[2]. Chemical separation of the ash samples is carried out using an anion exchanger resin (Biorad AG 1x 2) for isolation of Pu and a TRU-Spec, column (EiChrom) for subsequent extraction of Am. After electrodeposition onto stainless steel plates the Pu and Am fractions are analysed by means of high resolution alpha spectrometry[3]. For quality control, Lab 1 continuously participated in international intercomparison programs, e.g. organised by the German Federal Department for Public Health or the French 'PROCORAD' program.

The long-term retention of Am in the chest and the excretion of Pu in urine and faeces are shown in Figure 1. An interesting feature of this case is the time-dependent variation in isotope composition. This is due to two different effects. The inhaled material contained a high initial activity ratio for ^{241}Pu/^{241}Am of 75. Since ^{241}Pu has a relatively short half-life of 14.35 y and the decay product is ^{241}Am, there is a significant ingrowth of ^{241}Am. After 20 y and neglecting excretion the ingrowth of ^{241}Am yielded ~1.5 times the original amount of the deposited ^{241}Am.

Table 3. Evaluation of the inhalation case by different organisations.

Identification	Type of study	Model used
EIR, 1983	First EIR evaluation[4]	ICRP 26, ICRP 30
EIR, 1984	Second EIR evaluation[5]	ICRP 26, ICRP 30
NRPB, 1992	Chromosomal analysis on a blood sample[6]	
FZK, 1995	Comparison of different models[7]	FZK ^{241}Am model[8], ICRP 26, IRCP 60
PNL, 1995	Comparison of different models[9]	ICRP 48, ICRP 66, ICRP 67, Tancock 93[10]
NRPB, 1996	Evaluation of the inhalation case[11]	ICRP 66, ICRP 67
IAEA, 1999	Intercomparison project[12] with 25 participants	Various
FZK, 2000	Intercomparison project[13] with 50 participants	ICRP 30, ICRP 54, ICRP 66, ICRP 67
FZK, 2002	Optimisation of ICRP model[14]	ICRP 67 modified
ICRP, 2002	Application of the ICRP Human Respiratory Tract Model[15] with ^{241}Am data	ICRP 30, ICRP 66, ICRP 67
IDEAS, 2005	Intercomparison project[16] with 35 participants	ICRP 66, ICRP 67

Table 4. Results of the different evaluations.

Identification	Estimated committed dose (Sv)					
	^{238}Pu	^{239}Pu + ^{240}Pu	^{241}Am	Total alpha	^{241}Pu	Total
EIR, 1983				0.1–1.2		
EIR, 1984				1.3	0.2	1.5
FZK, 1995						
ICRP 26	0.74		0.13	0.87	0.23	1.10
IRCP 60	0.55		0.09	0.64	0.15	0.79
PNL, 1995				0.46		
NRPB, 1996	0.08	0.72	0.09	0.89	0.11	1.0
IAEA, 1999	GM: 0.11	GM: 0.63	GM: 0.14			
	AM: 0.13	AM: 0.88	AM: 0.21			
	(0.04–0.3)	(0.2–1.9)	(0.04–0.8)			
FZK, 2000						
ICRP 30 +		GM: 0.32				
ICRP 54		AM: 0.45				
		(0.07–1.4)				
ICRP 66 +		GM: 0.19				
ICRP 67		AM: 0.24				
		(0.04–0.6)				
FZK, 2002	0.044	0.28 + 0.13	0.13	0.58	0.058	0.64
ICRP, 2002			0.06			
IDEAS, 2005		^{239}Pu only				
		GM: 0.14	GM: 0.053			
		AM: 0.16	AM: 0.070			
		(0.001–1.1)	(0.00006–0.3)			
NRPB, 1992	Average dose to lymphocytes: 0.5–2					

GM: Geometric mean. AM: Arithmetic mean of all results of the intercomparison, minimum and maximum in parentheses

In addition early excretions through urine and faeces showed different behaviour. Lung clearance in the early days resulted in a nuclide composition of the faeces equal to the inhaled mix. Otherwise, early urine samples showed a high concentration of ^{241}Am since americium in lungs has a higher excretion rate via urine than plutonium. After several years the excretion is not dominated any more from direct deposition in the lungs and, therefore, the composition of the radionuclide in urine and faeces has changed over time and has become similar as shown in Figure 2.

THE INTERPRETATION OF THE MEASUREMENTS

This inhalation case has been analysed by quite a number of institutions. The early evaluation had to be done at PSI for reporting to the competent authorities[4,5]. Later on the scientific interest in the case came up, especially since retention and excretion were not influenced by chelating agents. The first international study of the case was performed under a CEC contract in the Third Nuclear Fission Framework Programme (1992–1995). In the following years the case was subject of numerous actions as shown in Table 3.

Intake was estimated by different institutions using various models. These estimates varied over more than an order of magnitude. Based on the ICRP Publications 66 and 67 the most probable values are 30–40 kBq of alpha emitting Pu isotopes and ^{241}Am, and 200–300 kBq ^{241}Pu. The wide variation of the estimates has shown, that intake is only then a meaningful quantity to characterise the extent of an inhalation case if the model used for intake estimation is given and the same model is used to determine committed dose.

The estimation of committed dose has shown a wide variation among different institutions. The reason for this seems to be the variability of the local application of procedures and methods of dose estimation and not primarily the metabolic and dosimetric models used as such. In the 20 y observation period of this case the recommended internal dosimetry models have changed mainly from ICRP 26/ICRP 30 to ICRP 66/ICRP 67, not to mention the many supporting documents and the various models proposed by individual scientists. Overall, the interpretation and the dose estimation of the case have not changed dramatically with the application of the new models. It is assumed that the effective committed dose (E_{50}) is in the order of 1 Sv with an uncertainty of <0.5 Sv.

CONCLUSIONS AND OUTLOOK

The 1983 Pu inhalation case has become an important issue for model validation studies and training events in internal dosimetry. It is planned to continue the measurements as long as the subject is willing to participate. Following this overview of the history of the inhalation case a scientific paper on the application of the actual ICRP models will be prepared in collaboration with colleagues from other institutions.

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ANNEX: RESULTS OF ACTIVITY MEASUREMENTS

Table A1. Chest measurements of ^{241}Am.

Laboratory	Time after intake (d)	Activity of ^{241}Am (Bq)
Lab 1	0.1	390
Lab 1	1	310
Lab 1	3	230
Lab 1	6	240
Lab 1	15	230
Lab 1	34	230
Lab 2	38	260
Lab 1	44	230
Lab 1	160	220
Lab 2	164	230
Lab 1	357	220
Lab 1	1077	240
Lab 1	2925	180

The ^{241} Am detected in the chest measurements at later times results from ^{241}Am located in the lung, the pulmonary lymph nodes and the ribs of the subject.

Table A2. Activity of ^{241}Am in lung, liver, lymph and bone.

Laboratory	Organ	Time after intake (d)	Activity of ^{241}Am (Bq)
Lab 3	Lung	3724	120
Lab 2	Lung	3828	120
Lab 2	Lung	5110	133
Lab 3	Liver	3724	57
Lab 2	Liver	3828	24
Lab 2	Liver	5110	17
Lab 3	Lymph	3724	26
Lab 2	Lymph	3828	72
Lab 2	Lymph	5110	76
Lab 3	Bone	3724	69
Lab 2	Bone	3828	65
Lab 2	Bone	5110	95

Table A3. Excretion in urine.

Lab	Time after intake (d)	^{239}Pu+ ^{240}Pu [mBq/d]	^{241}Am+^{238}Pu [mBq/d]
Lab 1	1	11	110
Lab 1	2	41	100
Lab 1	13	6	17
Lab 1	14	4	15
Lab 1	21	4	11
Lab 1	30	4	6
Lab 1	31	4	6
Lab 1	37	6	6
Lab 1	180	3	4
Lab 1	43	4	5
Lab 1	179	5	4
Lab 1	180	3	4
Lab 1	182	4	6
Lab 1	183	3	4
Lab 1	364	5	4
Lab 1	365	2	3
Lab 1	366	2	2
Lab 1	369	4	5
Lab 1	370	5	7
Lab 1	371	4	4
Lab 1	605	3	3
Lab 1	606	4	5
Lab 1	607	2	2
Lab 1	1074	2	2
Lab 1	1075	4	3
Lab 1	1076	3	3
Lab 1	1077	6	3
Lab 1	1921	6	6
Lab 1	1922	5	5
Lab 1	1923	7	4
Lab 1	2089	7	5
Lab 1	2090	5	4
Lab 1	2091	7	3
Lab 1	3902	3	3
Lab 1	5076	9	4
Lab 2	6043	11	6
Lab 2	6044	10	3
Lab 2	6045	16	6
Lab 1	6859	10	6
Lab 1	6860	10	4
Lab 1	6870	9	4
		^{238,239,240}Pu [mBq/d]	^{241}Am [mBq/d]
Lab 1	2092		2
Lab 1	2095		4
Lab 2	2525	4	<1.5
Lab 2	2526	6	5
Lab 2	2527	7	3
Lab 2	2921	5	3
Lab 2	2922	4	2
Lab 2	2923	<1.5	<1.5
Lab 3	3723	10	
Lab 3	3725	9	
Lab 2	6043	13	5
Lab 2	6044	10	2
Lab 2	6045	17	5
Lab 1	6859	10	5
Lab 1	6860	10	3
Lab 1	6861	9	3

Table A4. Excretion in feces.

Lab	Time after intake (d)	$^{239}Pu+ ^{240}Pu$ [mBq/d]	$^{241}Am+ ^{238}Pu$ [mBq/d]
Lab 1	1	5 200 000	1 500 000
Lab 1	2	3 000 000	740 000
Lab 1	3	440 000	74 000
Lab 1	13	670	160
Lab 1	20	960	190
Lab 1	21	480	110
Lab 1	30	670	120
Lab 1	37	250	78
Lab 1	43	260	74
Lab 1	44	160	44
Lab 1	179	590	120
Lab 1	180	470	100
Lab 1	182	310	93
Lab 1	183	310	63
Lab 1	365	340	81
Lab 1	366	150	26
Lab 1	369	440	110
Lab 1	370	180	37
Lab 1	371	180	41
Lab 1	605	300	85
Lab 1	606	300	80
Lab 1	607	170	59
Lab 2	6043	12	5
Lab 2	6044	13	5
Lab 2	6045	8	2
Lab 1	6859	4	1
Lab 1	6860	14	3
Lab 1	6861	7	3
		$^{238,239,240}Pu$ [mBq/d]	^{241}Am [mBq/d]
Lab 1	1074	70	18
Lab 1	1075	120	30
Lab 1	1076	65	17
Lab 1	1077	25	7
Lab 1	1921	90	13
Lab 1	1922	190	46
Lab 1	1923	100	15
Lab 2	2525	47	17
Lab 2	2526	17	5
Lab 2	2527	38	13
Lab 2	2921	5	<1.5
Lab 2	2922	13	6
Lab 2	2923	3	<1.5
Lab 3	3723	15	
Lab 3	3725	11	
Lab 2	6043	13	4
Lab 2	6044	14	4
Lab 2	6045	9	2
Lab 1	6859	4	1
Lab 1	6860	14	2
Lab 1	6861	8	2

Table A5. Ratio of americium to total α-emitting plutonium in feces and urine.

Days	Ratio $^{241}Am/(^{238}Pu+^{239}Pu+^{240}Pu)$	
	Feces	Urine
1	0.13	8.9
2	0.11	2.1
3	0.08	
13	0.11	2.96
21	0.09	2.58
30	0.08	1.26
37	0.13	0.80
44	0.12	1.16
181	0.10	1.02
369	0.10	0.93
606	0.13	0.96
1076	0.26	0.56
1922	0.19	0.62
2526	0.35	0.63
2922	0.43	0.51
6044	0.26	0.26
6860	0.22	0.38