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X-ray fluorescence analysis of lead in human skeleton in vivo

by LARS AHLGREN, M.Sc.,[1] KURT LIDÉN, Ph.D.,[1] SÖREN MATTSSON, Ph.D.,[1] and STIG TEJNING, M.D.[2]

AHLGREN, L., LIDÉN, K., MATTSSON, S. and TEJNING, S. X-ray fluorescence analysis of lead in human skeleton in vivo. *Scand. j. work environ. & health* 2 (1976) 82—86. The lead concentration in the skeleton of living man was measured by X-ray fluorescence analysis. Five former workers from a metal industry were studied. The mean lead concentration in their skeletons was estimated to be 62 μg/g with a standard error of $\pm$ 5 μg/g. A comparison with the "normal" skeletal concentrations of lead in people from southern Sweden showed the skeletal concentrations of the men studied to be about three to nine times higher.

Key words: X-ray fluorescence, in vivo measurements, lead, skeleton, metal industry.

X-ray fluorescence analysis is a well established technique used in determining the elemental composition of various laboratory samples. The introduction of solid state detectors with high energy resolution has made it possible to use this technique for in vivo studies (3). In in vivo studies the tissue volume is irradiated with X- or γ-radiation and the resulting characteristic X rays are measured with a Ge(Li) or a Si(Li) spectrometer. Except for the unique occurrence of a high iodine concentration in normal thyroid tissue (400—600 μg/g), the stable elements studied in living man by this technique have been administered as a tracer (2).

The present work introduces the X-ray fluorescence method for in vivo determination of the lead content in the skeleton of workers in the lead industry.

EXPERIMENTAL TECHNIQUE

A 20 mCi ^{57}Co source, mainly emitting 122 keV γ rays, was used for the generation of the characteristic X-radiation of lead. The collimator and the radiation shield consisted of high purity tin. Moreover, we eliminated the small contribution of characteristic X rays from lead impurities in the collimator by covering its opening with high purity gold.

A 16 mm (diameter) $\times$ 5.2 mm Ge(Li) detector was used (energy resolution FWHM = 750 eV at 75 keV; entrance window = 0.13 mm Be). The angle between the incident and measured radiation was 90°.

IN VIVO MEASUREMENTS

Five males were studied. They had formerly worked in a metal industry and

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Table 1. Lead concentration in five former workers from a metal industry.

Person	Age (a)	Time of employment in lead industry (a)	Time after retirement (a)	Lead concentration in blood		Lead concentration in skeleton, determined by in vivo X-ray fluorescence analysis (μg per g ± 1 SD)			
				Mean value during the last 5 years of employment (μg per g)	Value at the time of bone measurements (μg per g)	Forefinger		Tibia (left)	
						Right	Left	Position 1	Position 2	Position 3
ON	66	17	2	0.68	0.43	70 ± 12		46 ± 9	96 ± 13	77 ± 15
JB	71	23	4	0.65	0.41	90 ± 15		75 ± 10	84 ± 15	
MB	64	22	4	0.73	0.32	52 ± 9		70 ± 11	74 ± 12	45 ± 12
AS	67	27	1	0.63	0.51	30 ± 8	41 ± 8	51 ± 11	79 ± 17	
HL	66	22	0.5	0.68	0.42	43 ± 7	58 ± 9	34 ± 12	55 ± 12	
Mean value (± 1 standard error)	67	22	—	0.67	0.42	55 ± 8			66 ± 5	

had handled much lead. They had retired 0.5—4 years before the measurements (table 1). During a period of several years they had shown a high lead content in their blood.

X-ray fluorescence measurements were made at three to four sites on every man, namely, on the left and/or right forefinger (fig. 1), on the tibia 8 cm from the ankle (position 1), on the tibia at about 20 cm from the ankle (position 2), and, in two cases, on the tibia at about 28 cm from the ankle (position 3).

Fig. 2 shows a typical pulse height distribution from a measurement of a forefinger. The distribution is dominated by pulses due to the Compton scattering of 122 keV (and 136 keV) photons at about 90°. One can also recognize a peak due to

Fig. 1. Arrangement for the determination of lead in fingers by X-ray fluorescence analysis. The radiation shield, the ^{57}Co source (20 mCi), the detector-collimator with its gold-covered opening, and the Perspex holder for the finger are shown.

coherent scattering of primary photons and peaks from characteristic X rays from gold in the collimator. The small peaks from K_{α_1}- and K_{α_2}-radiation from lead in the forefinger are located in the valley between the K_α and K_β peaks from gold.

The effective registration time at each site was 15 min, but the real measurement time lasted 20—30 min because of the dead time of the multichannel analyzer.

RADIATION DOSIMETRY

In the present experiment the absorbed dose at the surface of the skin was estimated to be less than 800 mrad for a 30-min irradiation period. Since the irradiated area was very small, about 3 cm^2, the integral dose was insignificant in comparison with that of an ordinary X-ray examination.

RESULTS AND DISCUSSION

For all measuring sites on all five subjects a statistically significant net number of pulses was found in the K_{α_1} and the K_{α_2} regions of the registered pulse height distribution (table 2).

Measurements on finger phantoms filled with distilled water or measurements on one of the authors gave no net number of pulses in the corresponding region.

In order to estimate the lead content in the irradiated tissue seen by the detector, we measured known concentrations of lead in different phantoms for comparison.

The forefinger measurements were compared with measurements on cylindrical Perspex tubes (wall thickness = 1.5 mm) of various diameters filled with a known concentration of lead nitrate in a water solution. As expected, the counting rate in the K_α peaks varied considerably with tube diameter. This counting rate was proportional to the irradiated water vol-

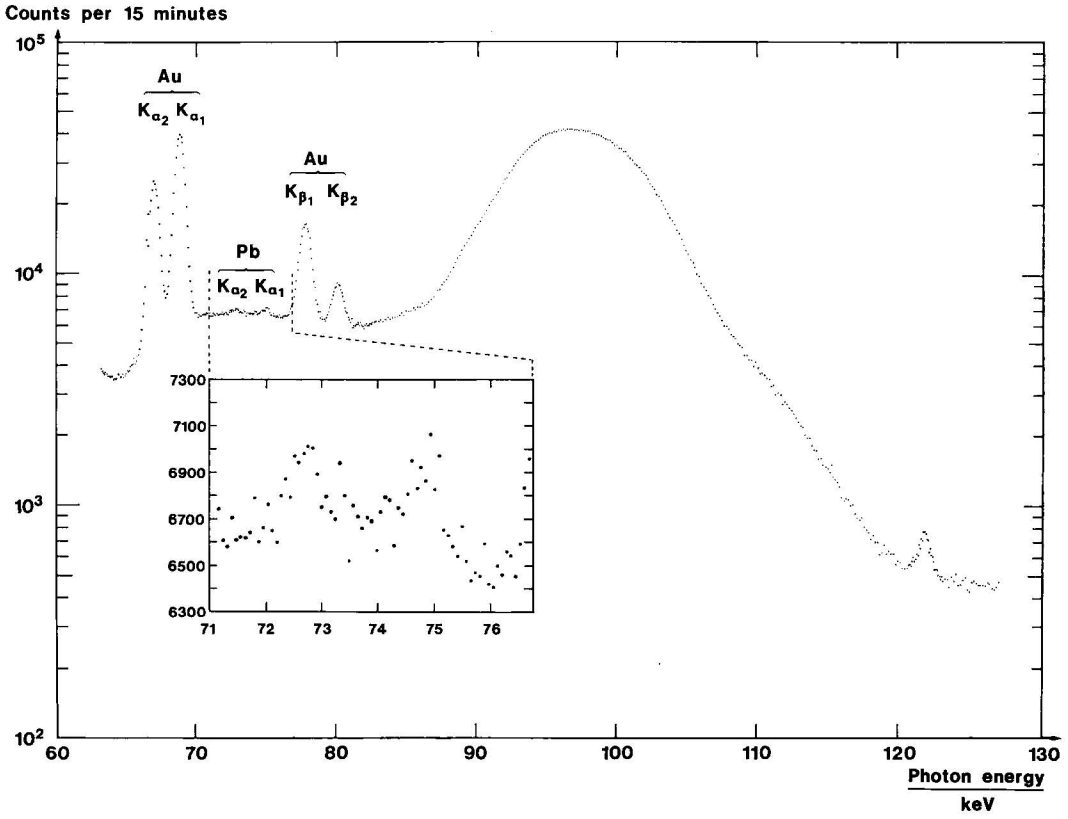

Fig. 2. Pulse height distribution recorded in an X-ray fluorescence study of the forefinger of a retired worker from the metal industry.

Table 2. Net counting rate (min^{-1}) in the Pb-Kα_1 and Pb-Kα_2 regions (74.97 ± 0.36 and 72.80 ± 0.36 keV).

Person	Net number of pulses per minute (± 1 SD)				
	Forefinger		Tibia		
	Right	Left	Position 1	Position 2	Position 3
ON	323 ± 33		287 ± 40	425 ± 35	251 ± 37
JB	305 ± 30		426 ± 47	278 ± 39	
MB	275 ± 34		386 ± 45	372 ± 45	186 ± 45
AS	161 ± 36	234 ± 34	251 ± 43	223 ± 36	
HL	253 ± 35	253 ± 32	145 ± 42	229 ± 42	

ume seen by the detector. For instance, if the inner diameter of the tube was increased from 15 to 25 mm, the counting rate increased by a factor of 2.4. In addition the counting rate in the Compton "peak" varied with tube diameter. If the outside diameter of the tube was increased from 15 to 25 mm, the counting rate increased by a factor of 2.2.

The number of Compton interactions is proportional to the electron density of the scattering material. As the number of electrons per mass unit is almost independent of the atomic number, the counting rate in the Compton "peak" is proportional to the mass of the volume examined. The attenuation of the incoming primary photons and the generated characteristic lead X rays is dominated by Compton interactions both in bone and in water. Therefore, as a first approximation, the ratio between the counting rates in the lead Kα peaks and the Compton "peak" is dependent only on the lead concentration and is independent of the atomic number and the density of the surrounding matrix.

In the finger measurements the "effective diameter" of the finger was estimated from the counting rate in the Compton "peak" of the phantom measurements. The relationship between counting rate in the Compton "peak" and the Kα peaks obtained from the finger phantom measurements was then used to estimate the concentration in the finger.

The tibia measurements were compared with measurements on cylindrical Perspex tubes filled with a known concentration of lead nitrate in a water solution and placed in a paraffin phantom. The ratio between the counting rate in the Kα peaks and the Compton "peak" was studied for the actual geometries and used for the estimation of the lead concentration in the tibia. The tibia measurements were less accurate than those of the forefinger mainly because of certain difficulties in defining the geometry. A comparison of the tibia and the finger measurements, however, strongly indicated that there could be no significant external contamination of the fingers.

Table 1 summarizes the estimated lead concentrations in the forefingers and tibia of the five persons studied. Table 1 also shows the blood lead content determined by atomic absorption spectrophotometry of blood samples obtained on the day the in vivo measurements were made. The mean value of the blood lead concentrations during the last 5 years of employment is also shown. According to table 1, on the average, no statistically significant difference in lead contamination between the forefinger and tibia was observed for the persons studied, and the individual lead concentrations did not differ significantly. The mean lead concentration determined for all the skeletal measurements, i.e., three to four sites on each of the five subjects, was 62 μg/g with a standard error of ± 5 μg/g.

According to the unpublished results of Schütz the "normal" blood lead concentration of people from southern Sweden is between 0.05 and 0.13 μg/g. Using a ratio estimated from Barry and Mossman (1), between the concentration of lead in bone and in blood for males between 60 and 79 years of age, we found the "normal" concentration in the skeleton of people of the same age from southern Sweden to be between 7 and 18 μg/g. Therefore the five men studied in this investigation had a lead content in the studied parts of their

skeleton which was about three to nine times higher than the calculated "normal" mean value.

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