$^{239+240}$Pu and heavy metals (Zn, As, V, Cu, Co, Ni, U, Pb, and Cr) in archived human samples from Finland

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A sample set of human livers, ribs, lungs and lymph nodes was analyzed for estimating concentrations of heavy metals and $^{239+240}$Pu in different organs. The samples were collected within autopsies made in Southern Finland and Lapland in 1976-1979. The sampling program was part of investigations concerning enrichment of radionuclides along the food chain lichen-reindeer-man (Jaakkola et al. 1981, Mussalo-Rauhamaa 1981). Furthermore, in this recent study, the enrichment of heavy metals in the food chain lichen-reindeer-man was investigated by determining their concentrations in reindeer and lichen samples.
Over 200 samples were collected from the autopsies of 67 persons during 1976-1979. In this preliminary study, 31 samples were used as test samples, both for

a) modifying existing analytical methods for biological samples and
b) to adjust the detection techniques (LSC, ICP-AES) for biological samples with high matrix content and low radioactivity concentration

The samples were ashed and/or wet-ashed with conc. HNO_3 and HCl. A fraction of this solution was taken for the determination of heavy metals by ICP-AES. From rest of the solution, $^{239+240}$Pu, ^{241}Am and ^{90}Sr were separated by ion exchange and extraction chromatography. The activity concentrations of $^{239+240}$Pu and ^{241}Am were determined with alpha spectrometry and that of ^{90}Sr with liquid scintillation counting.
The activity concentration of $^{239+240}$Pu per wet weight was 0.5±0.2 – 4.9±0.8 µBq/g in lungs (9 results), 21±5 µBq/g in livers (average of 2 results), 14±7 µBq/g in ribs (one result), and 9±3 µBq/g in collarbone (one result). The activity concentration of $^{239+240}$Pu in these human samples is in accordance with a previous study of the same sample collection (Mussalo-Rauhamaa 1981) and with worldwide values summarized by Taylor (1995). In most cases, the activity concentration of ^{238}Pu was close to or below detection limit and sometimes also $^{239+240}$Pu. This is a challenge for analytical method development, as large sample mass is needed and biological samples contain loads of interfering matrix (bone: calcium, lungs: iron, all: organic compounds). The composition of these samples was problematic for heavy metal determination as well, since some organic compounds were not destroyed during ashing+wet ashing procedure: iron-containing precipitation occurred in some final samples soon after diluting for ICP-MS-samples, and ICP-AES had to be used instead of ICP-MS for measuring heavy metal concentrations, due to better tolerance of former against sample impurities.
Heavy metal concentrations varied with orders of magnitudes inside and between sample categories (Table 1). Cobalt was not detected in any of the samples and lead was found only in 9 lung samples ($< D_L$ – 0.99±0.20 µg/kg wet weight). Elevated levels of lead, nickel and cadmium (not determined here) in lungs are typical for tobacco smokers. Average uranium contents for skeleton and soft tissues have been reported to be 3.79±0.45 µg/kg and about 0.5 µg/kg wet weight, respectively, for three Caucasian men (Kathren&Tolmachev 2015), our general uranium concentration being roughly at the same magnitude, but our values for ribs 0.18±0.04 – 0.46±0.09 µg/kg being lower than for the skeleton by Kathren&Tolmachev (2015).
Our highest value for nickel concentration in lungs, 3.74±0.75 µg/kg wet weight, is even higher than that of mine workers in Ontario, 1.84 µg/kg dry weight (Verma 2013). The same lung sample contained also the highest chromium content of lungs (14±3 µg/kg), as well as high amount of uranium (5.9±1.2 µg/kg), but lead content was low (0.28±0.06 µg/kg), indicating smoking as an improbable nickel source of this lung. This lung donor of our study lived 50 years in Sodankylä, Lapland, working as a reindeer herder outdoors most of the time. The lungs of this person might have exposed to pollutants from Kola Peninsula metal smelters via inhalation.
After this preliminary sample set and analytical method adjustment, the work for analyzing the rest of the human samples will continue and a larger data set will be produced.

Table 1. Heavy metal concentration in different human organs determined with ICP-AES (µg/kg wet weight).

Sample type (n)	Zn	As	V	Cu	Ni	U	Cr
lung (16)	< D_L – 52±10	< D_L – 0.11±0.02	1.30±0.26 – 13±3	< D_L – 1.96±0.39	< D_L – 3.74±0.75	0.68±0.14 – 7.3±1.5	< D_L – 14±3
lymph nodes (7)	< D_L – 29±6	< D_L – 0.07±0.01	3.02±0.60 – 45±9	< D_L – 6.8±1.4	< D_L – 1.59±0.32	< D_L – 3.39±0.68	< D_L – 2.59±0.52
rib (3)	< D_L – 15±3	-	3.06±0.61 – 4.12±0.82	-	-	0.18±0.04 – 0.46±0.09	-
flesh (2)	88±18 – 165±33	-	1.51±0.30 – 1.74±0.35	1.63±0.33 – 2.23±0.45	< D_L – 0.48±0.10	0.97±0.19 – 2.21±0.44	< D_L – 0.13±0.03
collar-bone (1)	99±20	0.04±0.01	1.90±0.38	1.88±0.38	1.36±0.27	2.71±0.54	0.24±0.05
liver (2)	79±16 – 141±28	-	2.41±0.48 – 5.2±1.0	6.4±1.3 – 7.9±1.6	1.31±0.26 – 1.61±0.32	2.89±0.58 – 4.52±0.90	-

References

T. Jaakkola, M. Keinonen, M. Hakanen, J.K. Miettinen. Actinides in Man and Animals. Proceedings of the Snowbird Actinide Workshop (M.E. Wrenn, ed.). RD Press, USA (1981), p. 509.

Helena Mussalo-Rauhamaa. Accumulation of plutonium from fallout in southern Finns and Lapps. Report Series in Radiochemistry 4/1981. Helsinki 1981.

D.M. Taylor. Environmental Plutonium in Humans. Applied Radiation and Isotopes 46, 1245-1252 (1995).

R.L. Kathren, S.Y. Tolmachev. Natural Uranium Tissue Content of Three Caucasian Males. Health Physics 109, 187-197 (2015).

D.K. Verma. Metals in the Lungs of Ontario Hardrock Miners. Archives of Environmental & Occupational Health 68, 180-183 (2013).