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Low Temperature Irradiation Applied to Neutron Activation Analysis of Mercury in Human Whole Blood

DAG BRUNE

AB Atomenergi, Studsvik, Sweden

The distribution of mercury in human whole blood has been studied by means of neutron activation analysis.

During the irradiation procedure the samples were kept at low temperature by freezing them in a cooling device in order to prevent interferences caused by volatilization and contamination.

The mercury activity was separated by means of distillation and ion exchange techniques.

The determination of mercury in small quantities has in recent times attracted increasing attention. Mention may here be made of, for example, cases of poisoning in connection with seed treatment with fungicides.[1]

Several methods for the determination of mercury in biological material by means of neutron activation analysis are now available.[2–6] In this connection separation techniques based on distillation, extraction, exchange reactions, precipitation and electrolytic deposition are employed, which may permit an accurate determination of mercury.

However, the analysis is still encumbered with certain problems attributed to the volatility of various mercury compounds. Thus, considerable losses of mercury during the drying of organic matter have been reported.[3,7,8] The irradiation of wet biological samples is otherwise connected with certain disadvantages. The pressure built up by radiolysis in the sealed ampoules may give rise to losses of volatile compounds when the ampoules are opened if the pressure is not decreased by cooling in, for example, liquid nitrogen before opening.

Losses of mercury due to volatilization from aqueous solutions (*e.g.* biological fluids or water), may be even more pronounced. This would occur, for example, during an evaporation performed in order to concentrate the sample before activation and also during the subsequent irradiation step. Thus, from solutions containing chloride or sulphate ions in concentrations of 100 ppm, volatilization losses of more than 50 % have been observed by the author when heating such solutions at temperatures of about 60°C for 12 h. This temperature is almost the same as that which arises in aqueous samples in irradiation positions generally applied for such samples in the reactor R1, Stockholm.

Table 1. Mercury content of human whole blood, expressed in μg mercury per g whole blood. Median value: 0.011 μg/g. Range: 8.

Sample	Age (y)	Sex	Mercury-content (μg/g)	Sample	Age (y)	Sex	Mercury-content (μg/g)
1	41	♀	0.003	11	39	♂	0.011
2	43	♂	0.003	12	34	♂	0.011
3	21	♂	0.004	13	35	♂	0.013
4	31	♀	0.006	14	35	♀	0.014
5	41	♂	0.008	15	37	♂	0.015
6	37	♂	0.008	16	37	♂	0.017
7	31	♂	0.010	17	40	♂	0.018
8	30	♂	0.010	18	29	♂	0.020
9	31	♂	0.010	19	37	♂	0.023
10	29	♂	0.011	20	49	♂	0.024

During the irradiation procedure, reactive species are formed in water producing for example, hydrogen peroxide which may, moreover, contribute to the volatilization processes, though probably not to any appreciable extent.

However, if the activation is performed at a low temperature by freezing the samples in a cooling device during neutron irradiation, these losses, as well as the contamination of the samples from the container walls, can be avoided.[9] In this way the accuracy of the analysis may be improved.

Low temperature irradiation has been applied in the present investigation to the determination of the mercury content of human whole blood. Using this technique the mercury content in such samples may be determined with a standard deviation of a single value of 7 %.[9]

EXPERIMENTAL

Blood samples of about 2 ml were collected in polyethylene containers from 20 healthy blood donors. The samples were irradiated in a thermal neutron flux of about 2×10^{12} n/cm^2 sec in R1, Stockholm for 13 h at a temperature of about -40°C in a cooling device described elsewhere.[9] After the "cold" irradiation procedure, the samples were allowed to melt for a short time in order to remove a thin surface layer which may have been contaminated by the container surface. The samples were then transferred to a closed system and destroyed with H_2SO_4 and H_2O_2. The mercury activity was then separated by means of distillation and ion exchange techniques before gamma-spectrometric measurements being undertaken.[5,]* The analysis was assayed by means of the nuclide ^{203}Hg.

RESULTS AND DISCUSSION

The amounts of mercury in the 20 samples of human whole blood investigated are given in Table 1. These values are nearly consistent with a skew distribution and the median, amounting to 0.011 μg/g, has consequently been chosen to represent the central value. The range of the mercury concentration, expressed as the ratio of the highest and lowest values, corresponded to a factor of 8.

* Yield: 96 %

The concentration of various elements in trace quantities in mammalian tissues, moreover, often exhibits a skew distribution.[8,10] The mercury content of human heart material also showed such a distribution, with about the same range as that obtained in the present investigation.[8]

The median value obtained for the mercury content in whole blood is about twice the mean value previously reported by Comar *et al.*[4] who also used neutron activation analysis in the investigation.

At concentrations down to 0.05 ppm, mercury can also be determined using non-radioactivation methods.[7]

Stock, who also examined the mercury content of human whole blood, applied a method based on microscopic observations.[11] In his investigation, a mean value was obtained which was nearly equal to the median value of the present investigation.

In Stock's work, the influence of the amalgam content of teeth on the mercury concentration of the blood was also studied. The presence of amalgam did not, however, seem to affect this concentration. On the other hand, the mercury content in various organs was appreciably increased in the case of persons with amalgam fillings.

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Appendix. In the determination of mercury in water, the freezing technique was found to be suitable in combination with the separation technique involving exchange reactions described by Kim and Silverman.[2] However, this separation technique was slightly modified by increasing the exchange time two-fold (10 min) and the mercury content four-fold (0.2 ml), resulting in a recovery of 98 % with a standard deviation of a single value of 1 %.

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