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Original Research

Mercury Content in Rural and Industrial Regions in Lower Silesia

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Abstract

Monitoring studies of environmental (soil, water, grass and cereals) and biological materials (tissues of hens, eggs, cow's milk) in the industrialized area of the Legnica-Głogów copper mine district (LGOM-copper region) and Jeleniogórska Valley (KJG) were carried out in order to determine the content of mercury. Low concentrations, at levels naturally occurring in the environment, were determined. The content of this metal in soil, plants, water, poultry tissues and raw cow's milk was slightly elevated in both the industrial (LGOM-copper region) and ecologically clean (KJG) regions.

Keywords: mercury, environment, hen, egg, milk, Lower Silesia

Introduction

Mercury is abundant in the environment. In air in agricultural regions, it is most frequently at a level of 2-6 $\mu g \cdot m^{-3}$ and in large agglomerations the content lies within the range 5-50 $\mu g \cdot m^{-3}$. In waters of natural rivers the amount is 0.002-0.007 $\mu g \cdot dm^{-3}$, and in polluted waters the concentration can be as high as 1.2 $\mu g \cdot dm^{-3}$ [1]. The content of mercury in soil may reach as high a level as 5 $\mu g \cdot kg^{-1}$ d.m. in regions of mines and chemical industry impact, but the average level is 0.1-40 $\mu g \cdot kg^{-1}$ d.m. [2-4]. In cultivated plants it may reach different levels, depending on the concentration in the soil and in the air. In cereal grains mercury content averages 4-21 and in grasses the content is as high as 68 $\mu g \cdot kg^{-1}$ s.m. [1, 5].

In tissues and organs of animals, as well as in animal products (milk, meat, fat, eggs, honey), various concentrations of this element are reported, e.g. in hen eggs from backyard husbandry the maximum content of Hg was 13.66, in hen muscles 8.55 $\mu g \cdot kg^{-1}$ and in hen livers 11.46 $\mu g \cdot kg^{-1}$ [6, 7]. In cow's milk from the polluted regions, the concentration of Hg reaches the maximum level of 6.7 $\mu g \cdot kg^{-1}$ [8]. In some regions of EU, the elevated level of Hg in eggs, meat and giblets, as well as in milk products was recorded [9].

Some authors report that in wild animals (roe-deers, hares, wild boars), relatively high concentrations of Hg may be present in kidneys and liver [10, 11]. Also, an elevated concentration of Hg was reported for various fish species from the Baltic Sea and also living in inland waters [12, 13].

The toxicity of mercury depends on its chemical form. The most harmful were found to be metal-organic forms, which are first of all methylmercury, but also phenyl-, ethyl- and methoxyethyl-mercury compounds. Methylmercury is the primary form which is of concern when considering human and animal health because it is readily bioaccumulated and biomagnified. This form moves easily among the

Table 1. The type and number of analytical samples from KJG and LGOM regions.

Material	KJG		LGOM	
	No. of farms	No. of samples	No. of farms	No. of samples
Drinking water	8	8	8	8
Soil from yard	8	8	8	8
Grain cereals	8	12	8	11
Grass from yard	8	8	8	8
Meadow hay	8	8	9	9
Laying hen	8	8	8	8
Hen's egg	8	8*	8	8*
Cow's milk	8	13**	9	19**

*- 1 sample – average from 3 eggs.
** - 1 sample – average from milking from 4 quarters of udder.

levels of the trophic chain. Methyl mercury is also formed from other sources of mercury (originating from burning of coal, industrial processes, etc.) by bacterial conversion to organic methyl mercury (biomethylation process). Compounds of mercury impair almost all enzymatic reactions, including the processes of protein biosynthesis, and pose pathological changes in the nervous system (in particular in the brain), causing even death in extreme cases [14].

According to Zahir et al. [15] and Virtanen et al. [16], mercury was found to be an important factor causing a variety of malfunctions and metabolic disorders in humans, such as neurological, nephrological, immunological, cardiological, motoric, fertility and genetic.

The source of mercury emissions to the environment is the combustion of fuels, mainly bituminous coal, brown coal, and the iron and steel industries, as well as non-ferrous metallurgy, petroleum processing, cement production, and also from other industrial processes which employ the use of mercury and its compounds, the combustion of wastes, wastewater treatment plants and others. For instance, various types of coal contain 0.01-1.8, municipal wastewater 8.8-9.5 and ash from municipal wastes contains 31.4 mg $Hg \cdot kg^{-1}$. Limestone contains an average of 0.414, gypsum - 0.116, and cement 0.060 mg Hg/kg [1, 17-19].

In the 1980's, the estimated value of annual global Hg emissions to the atmosphere was 8,000 Mg and in the late 1990's it was estimated at 6,000 Mg/year [20]. Recently, the tendencies have not changed, although the concentrations tend to slightly decrease [21].

Mniszek and Zielonka [18] have reported that the main source of Hg emissions to air in Poland are the power coal and cement industries. The subsequent emitters include metallurgy, chlorine production and spent mercury lamps. The authors calculated that in 1992, mercury emission from the main sources was 35.88 Mg/year in Poland. The real value is probably much higher, since the other sources of Hg emission were not taken into consideration, such as: the production of pigments, paints, pharmaceuticals, cells, asphalt, glass, petroleum refinery, textile industry, the production of fertilizers, combustion of gases and liquid fuels. At the beginning of the current decade (the year 2000), the emission of Hg in Poland from power plants and industry was estimated at 25.6 Mg/year, with a decreasing tendency in the subsequent years [22].

Mercury in Poland poses a serious toxicological problem and it is easily distributed, which can lead to local increased accumulation of this toxic metal in rural environments, in organisms of plants and animals.

The aim of our present monitoring work was to assess mercury content in the rural environment, in elements of the environment as well as in biological material in a non-polluted region (Jeleniogórska Valley) and in the Legnica-Głogów Copper Mine District industrialized area (LGOM-copper region).

Experimental Procedures

Monitoring studies were carried out in 16 small-scale farmsteads in the Jeleniogórska Valley region and in LGOM-copper region, from the following communes: Rudna, Polkowice and Grębocice, where plant cultivation and livestock husbandry is undertaken on a small scale (Table 1). Samples of feed (grain of cereals and meadow hay), soil (top layer) and grass from courtyard and drinking water were taken from the selected farms.

From a single laying hen, randomly selected from each farm, samples of muscles (thigh), livers, kidneys and cerebrum were taken after decapitation. Also, 3 eggs were selected from each farm, which were crushed with the separation of yolk, albumen and shell. Milk was sampled directly from cows, from afternoon (evening) milking, from all teats in similar proportions. The material was placed in sterile plastic containers and transferred to an analytical laboratory. Total mercury was determined in environmental (soil, water, plant) and biological (poultry, eggs) material, after previous mineralization. The analysis was carried out by atomic absorption spectrometry (AAS), flameless amalgamation technique with the use of TMA-254 analyzer (the lowest detectable quantity – 0.2 ng). Certified reference materials DOLT-2 and DORM-2 (Canadian Certified Reference Material Project) were used.

The determination of Hg in cow's milk (raw) was performed using atomic absorption spectrometry (AAS) by an AMA-254 mercury analyzer (the lowest detectable quantity of Hg – 0.1 ng). The certified reference material used was Non-Fat Milk Powder (U.S. Department of Commerce).

The results were elaborated statistically. The average values were calculated ($\bar{x}$), standard deviation (SD), percentage and statistical differences between the average content of mercury in the studied groups were determined using t-test (Microsoft Excel 2000 software).

Table 2. Comparison of Hg content in environmental and plant material from the LGOM and KJG areas (μg/kg).

No.	Material	Region LGOM		Region KJG	
		x ± s	range	x ± s	range
1.	Drinking water	1.20 ± 1.45	0.3 - 4.4	0.81 ± 0.42	0.4 - 1.7
2.	Soil from yard	408a ± 168	234 - 607	232 a* ± 172	117 - 572
3.	Grain cereals	5.14 ± 1.49	234 - 667	3.96 ± 3.18	1.0 - 10.9
4.	Grass from yard	38.8 ± 23.3	4.0 - 6.9	29.9 ± 19.1	19.0 - 53.3
5.	Meadow hay	18.5 ± 4.4	8.3 - 45.0	16.2 ± 5.24	6.5 - 38.6

a, a* -significantly different from soil in Region LGOM by a t-test $p<0.05$.

Table 3. Comparison of Hg content in biological materials (animal origin) from the LGOM and KJG areas (μg/kg).

No.	Material	Region LGOM		Region KJG	
		x ± SD	range	x ± SD	range
1.	Hen- muscle	2.14 ± 1.19	0.8 - 5.0	2.40 ± 0.75	1.2 - 4.0
2.	Hen - liver	23.4 ± 18.2	41.0 - 55.3	34.1 ± 66.6	2.1-196.6
3.	Hen - brain	5.95 ±2 .24	3.2 - 10.3	3.61 ± 2.46	1.1 - 7.9
4.	Hen – kidney	4.60 ±1 .86	3.0 - 8.2	3.58 ± 1.94	1.1 -6.5
5.	Egg - yolk	5.19a ± 2.09	2.5 - 8.2	3.09a* ± 1.23	1.3 - 4.6
6.	Egg- protein	3.29a ± 1.57	1.8 - 6.4	1.68a* ± 0.86	0.6 - 3.2
7.	Egg - shell	2.40 ± 0.95	0.8 - 3.7	1.87 ± 0.57	0.9 - 2.9
8.	Cow's milk	1.65 ±0 .52	0.8 - 3.5	1.33 ± 0.56	0.8 - 2.3

a, a* -significantly different from soil in Region LGOM by a t-test $p<0.05$.

Results and Discussion

The results of measurements of the content of mercury are presented in Tables 2 and 3. The average levels of Hg in the studied environmental (soil, grass, water, feed) and biological samples (muscles, livers, kidneys, cerebrum, eggs, milk) differed significantly.

In LGOM region, the content of Hg in drinking water from 7 towns ranged between 0.3-4.4 μg/kg, but in soil from rural courtyard was in the range 247-667 μg/kg. However, in KJG region the concentration of Hg in drinking water from 8 towns ranged between 0.4-1.7 μg/kg and in soil was within the range 117-572 μg/kg. The average concentrations of Hg in LGOM region were 48% higher for water and 76% higher for soil. But beyond this, the differences were not significant since the variability in the groups was high (high standard deviation).

In natural river waters, mercury is usually present at a level of 0.002-0.007 $\mu g \cdot dm^{-3}$, but in pollution the concentration reaches 1.2 $\mu g \cdot dm^{-3}$. Polluted underground waters were in the range 0.5-4 μg Hg/l [1]. The permissible concentration of Hg in drinking water is 1.0 μg/l [23]. In the studied samples of drinking water from 15 farms in LGOM and KJG, the concentration exceeded the recommended level on 4 farms (26.6%).

The concentration of mercury in soils was found to lie within a wide range. Chojnacka et al. [2] reported that in some soils of LGOM, mercury was present at a level of 0.57-4.64 mg/kg, Bontidean et al. [24] reported Hg concentration in soils of vegetable gardens in Denmark (0.02-37.79 mg/kg), but in a mining region in Portugal soil contained 109 mg/kg [25] and in Slovenia from 0.3 to as high as 973 mg/kg [26]. Taking into consideration the data mentioned above, we concluded that the results from 15 farms in LGOM and KJG pointed to on elevated level in only 3 cases (20%), which exceeded the recommended concentration of 0.5 mg/kg. Bieszczad and Sobota [5] report that a concentration of Hg in soil in the range 0.3-5 mg/kg d.m. is toxic to plants. Florencka and Chmiel [27] showed that a concentration of 0.1 mg Hg/kg in soil resulted in an 85% reduction of mesophilic bacteria. This concerned mercury oxide and mercury chloride. Mercury nitrate reduced bacteria count in the range 42-90%.

A different range for the content of Hg was obtained in courtyard grasses and hay, as well as in cereal grain, feed for livestock. In LGOM, the content of Hg in the grains of cereal from 7 towns ranged between 2.8-6.9 μg/kg, in grasses 10.4-75.4 μg/kg and in meadow hay 8.3-45.0 μg/kg. However, in the KJG region, the concentration of Hg in grains was 1.1-10.9 μg/kg, in grasses 19-53.3 μg/kg, and in

meadow hay 6.5-38.6 μg/kg. The average concentrations of Hg in LGOM were thus higher when compared with KJG, but the differences were not statistically significant, since large variability in groups was observed (high standard deviations, in particular in relation to grasses).

In the cultivated plants, the concentration of Hg may reach different values, depending on its concentration in the soil and in the air. Also of importance is the type of soil and its pH, the duration of vegetation season, etc. [1, 5].

When considering the above data, it is worth mentioning that data obtained from 16 farms in LGOM and KJG pointed to an elevated level of Hg in grasses, which could be explained by the multiplicity of functions they play in rural farms [5, 28]. The acceptable content of Hg in feeds and feed materials is 0.1-0.5 mg/kg with the maximum content of water 12% [29].

Mercury from soil, as well as plants from rural courtyards, rainwater, and various organic and mineral wastes, is accumulated by poultry in backyard husbandry. This may result in excessive bioaccumulation of this element in organisms of birds as well as in eggs laid by them.

In tissues, organs and eggs of hens, variable concentrations of Hg were determined. In the LGOM region the values ranged in thigh muscles 0.3-5.0, in liver 2.7-55.3, kidneys 3.0-8.2, cerebrum 3.2-10.3, egg yolk 2.5-8.2, egg albumen 1.8-6.4, egg shell 0.8-3.7 μg/kg fresh mass. However, in the KJG region in muscles the level was within the range 0.9-3.7, in liver 2.3-196.6, kidneys 1.1-6.5, cerebrum 0.9-7.9, egg yolk 1.25-4.6, egg albumen 1.0-3.2, and egg shell 0.95-2.9 μg/kg fresh mass. High content of Hg in two samples of liver in KJG and cerebrum of hen in LGOM is worth underlining.

The average values for Hg were slightly higher in livers and muscles of hens from KJG when compared with LGOM, but the average concentrations in kidney, cerebrum and eggs were slightly higher in LGOM when compared with KJG. The differences were not significant (beside egg yolk and egg albumen), since high variability in groups was observed (high standard deviations, in particular in the case of liver).

In the available literature there are many works on accumulation of mercury by poultry. Niespodziewański and Wierciński [30] investigated the content of Hg in 10 anatomic elements of young Polbar hens. The highest level of Hg (mg/kg d.m.) was detected in heart (0.309) and liver (0.0261) and the lowest in fatty tissue (0.0037) and in gizzard (0.0071). Opaliński et al. [7] found that the highest concentration of mercury contained livers of hens kept in an industrialized region – averaging 4.80 $\mu g \cdot kg^{-1}$, and maximally even 11.46 $\mu g \cdot kg^{-1}$, then lungs (averagely 3.66) and muscles (averagely 2.89). In an agricultural region slightly higher was the content of Hg in liver, but higher in lungs when compared to an industrial region. At the end of the 1980s, Falandysz [31] monitored the concentration of Hg in animals decapitated in northern Poland, including poultry. The value 5-12 $\mu g \cdot kg^{-1}$ was reported.

Żmudzki et al. [32] reported the content of Hg in muscles of hens kept in small-scale farms and large farms on the level of 5.0 and 3.0 $\mu g \cdot kg^{-1}$ f.m., respectively, but in livers 9.0 and 8.0 $\mu g \cdot kg^{-1}$ f.m., respectively. The data are the average for 450 hens from the whole country from the beginning of the 1990s. Żarski et al. [33] found that in healthy broiler chicks, the concentration of Hg in liver was 0.019, kidneys 0.068, thigh muscle 0.030, and cerebrum 0.005 mg/kg fresh mass.

There are only a few reports on the concentration of mercury in poultry eggs. Dobrzański et al. [34] reported that in egg content of backyard poultry keeping, litter and battery keeping, the average levels of Hg were as follows: 8.55, 1.02 and 1.33 $\mu g \cdot kg^{-1}$, respectively. In the case of eggshells, the concentrations were as follows: 7.77, 1.06 and 127 $\mu g \cdot kg^{-1}$. Other papers report [35] that the average level of Hg in duck's egg content in industrially polluted regions averaged 137.2 $\mu g \cdot kg^{-1}$ f.m, and typically agricultural regions (ecological) only 39.7 $\mu g \cdot kg^{-1}$ f.m. In the case of the content of duck's eggs the values were 55.1 and 10.5 $\mu g \cdot kg^{-1}$ f.m., respectively. The highest quantities of Hg were accumulated by egg yolks and the lowest by egg shells. It is also worth mentioning the study of Heinz and Hoffmann [36]. In wild ducks, the concentration of Hg in egg content increased to the maximum values of 35 mg/kg f.m., if fed with feed containing 20 mg/kg Hg in the form of methylmercury.

When taking these data into consideration it can be concluded that the obtained results of the content of Hg in tissues, organs and eggs of hens from backyard poultry keeping were in some cases elevated and exceeded the allowable limits, in particular for liver.

According to the Directive of the Polish Minister of Health from Jan. 1, 2003 [37] the allowable content of mercury (NDS) in meat of mammals and poultry was 0.02 and in other animal products 0.01-0.03 mg/kg f.m. depending on the content of dry mass. For liver and kidney the limit was 0.5 mg/kg f.m.

The available literature includes many reports on the accumulation of mercury in cow's milk, from cows bred in industrial and in typically agricultural regions. Dobrzański et al. [8] found that the content of Hg in milk of cows from various regions of the Silesian macroregion are within the range 0.3-6.7 mg/l. In the late 1980s Żmudzki et al. [38] found that Hg in cows milk from the Zgorzelec-Bogatynia region, average 2 mg/kg (max. 8 mg/kg). The acceptable limit for Hg concentration in milk – used as a component of food – was officially assessed at 10 mg/kg [37].

In Spain (Canary Islands) in consumption milk, the average concentration of Hg was determined at 0.09-0.61 mg/kg [39], but in China it is 1.0-3.9 mg/kg [40]. In Italy, in the vicinity of Rome, the concentration was significantly higher at 0.9-38 mg/kg, but in the Southern region of Italy the content was 2-3 mgHg/kg d.m. [41]. High concentrations were identified in milk products in Egypt 86-556 mg/kg [42]. Taking this into consideration, it's interesting that milk of women may contain from 0.2 to 20.3 mg Hg/l [43, 44].

Generally, it is thought that Hg transport to blood or milk from respiratory or digestive systems depends on the form of Hg, since only alkyl compounds of this element (eg. methylmercury) are easily transported. However, inorganic forms are excreted with excrements (urine, feces).

Grega et al. [45] report that Hg as well as other metals (Al, Pb, Zn) and S are first of all deposited in various organs (liver, kidneys, cerebrum) and afterwards can be transported from blood to milk, and mammary glands itself play a role of biological barrier [43]. It is known that Hg may form many persistent complexes with proteins and other compounds, mainly those which contain SH groups. According to Sundberg et al. [44], casein, fat and whey fraction of milk of mice accumulate mercury from methylmercury in percentage quantities 11:39:34; but from inorganic forms in the proportions 31:15:41 (%), respectively, as related to the total content of Hg in milk of these animals.

The presence of Hg in feed and drinking water, and to a lower extent in air, may influence the elevated content of Hg in milk. Chodorowski et al. [46] reported high concentrations of Hg in milk (23 mg/kg) if feed was contaminated with metallic mercury. However, Dobrzański and Tarnacka [47] showed that the content of Hg in milk of cows in the vicinity of waste dumps from the electro-technical industry averaged 3 mg/l. In pasture grass the maximum values reached as much as 1730 mg/kg d.m., which confirms that inorganic forms of this element are bioavailable only to a small extent.

In conclusion, in Lower Silesia there is no danger of contamination with mercury compounds caused by their presence in particular elements of the environment, as well as in biological material, which points to only the presence of natural or slightly elevated forms of this metal.

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