Original Research

Assessing Mercury Content in Plant and Animal Raw Materials in an Area Impacted by the Copper Industry

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Abstract

We used a mercury analyzer to determine total mercury (Hg) content in samples of plants and animal origin raw materials collected from an area impacted by the copper industry (Legnica-Głogów Copper District). In 2014 and 2015 we analyzed samples of wheat (grain); potatoes (tuber); dry grass (hay); cow milk, hair, and blood; and poultry liver, muscles, and eggs from 14 villages. Generally, no increased Hg concentrations in the analyzed samples were found. There were no significant differences between the examined years. The highest content of Hg was found in grass (hay) and cow's hair. There was a many-fold decrease of mercury concentrations in cereals, cow milk, and poultry (eggs, muscles, liver) in comparison to previous studies (2002-07), which indirectly proves that the environmental efforts led by local authorities and industry have been effective.

Keywords: copper industry, total mercury, plant, livestock

Introduction

Mercury belongs to the group of toxic metals and it exhibits strong chemical and biological activity and variability of its form (liquid and gas). It is not needed in the life processes of plants, animals, and humans. Mercury is toxic to the environment – especially in its organic forms (methyl mercury, ethyl mercury, phenyl mercury, etc.) [1-3]. The phytotoxic and genotoxic activity of Hg (and its compounds) is well documented [4-6], and the International Agency for Research of Cancer (IARC, Lyon, France) qualified organic mercury (methylmercury) compounds to group 2B, meaning that they are possibly carcinogenic to humans [7].

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Mercury is transferred between air, soil, and plants, and originates from several sources. Natural Hg sources include cannabar (ore) and fossil fuels such as coal or petroleum, as well as volcano eruptions and forest fires. Mercury content in the environment has been increasing due to discharge from hydroelectric, mining (mainly small-scale gold refining and processing), pulp and paper industries. Incineration of municipal and medical waste and emissions from coal-burning power plants also contribute to high levels of mercury [8-9].

It is difficult to evaluate the total global annual emissions of mercury with high precision, both from natural and anthropogenic sources, but it was estimated at the end of the 20th century on the level of between 4,400 and 7,500 Mg per year [10]. According to other data, global mercury emission to the atmosphere in 2010 only from major anthropogenic sectors (including artisanal and small-scale gold mining, coal combustion, non-ferrous metals, cement production, etc.) was between 1,010 and 4,070 Mg [9]. In Poland, according to different scenarios, Hg annual emissions in 2020 will be between 8.2 (MFTR restrictive scenario) and 21.6 Mg (SQ scenario), while for the base year 2005 it was estimated to be 21.2 Mg [11]. The mercury emission forecast from 10 European countries (including Poland) with the largest emission assumes its reduction to 179.6 Mg per year (BAU scenario) or 64.4 Mg per year (DEG scenario) in 2020 [12-13]. The issue of mercury is of particular interest in the European Union, thus a special CONTAM panel of EFSA (European Food Safety Authority), which is monitoring contaminants in the food chain, is currently dealing with the Hg emissions problem [14]. In September 2014, Poland, along with more than 140 other countries, signed the Minamata Convention, which seeks to reduce and ultimately ban the use of mercury and its compounds. The emphasis was primarily put on the prohibition of metallic mercury use in gold mines, but by 2020 this ban shall include all products containing mercury and its toxic compounds. For now, the ban does not include thimerosal, which is still used in some vaccines and cosmetic products. This does not mean that mercury will disappear completely. Certain compounds of this element will continue to be used in industry as catalysts as well as in commonly used technologies and scientific research [3, 8, 15].

In Poland, the limits of mercury and its compounds content have been established for atmosphere [16], workplace [17], drinking water, soil, and feed [18-20]. EU limits have been established for food, but only for fish and fish products [21]; however, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a weekly intake of methylmercury compounds from all sources tolerable by healthy human organisms (Provisional Tolerable Weekly Intake, PTWI), and it is 1.6 µgHg/kg body weight (BW), which corresponds to 0.096 mg per adult (60 kg weight), while the PTWI value for inorganic mercury is 4 µg/kg BW (0.24 mgHg/adult) [22].

The main sources of mercury in humans are marineorigin products, especially fish and fish products [2325], although some quantities may come from animal production (milk, meat, eggs) – especially in industrial areas, or from animals fed with feed contaminated with mercury compounds [26-29]. Tables 1 and 2 show data concerning mercury content in various environmental components (air, soil, water, fuel, fertilizers, sewage, etc.) and biological material (plants, feed, animals, food) according to various foreign and Polish authors.

The effect of the copper industry on the environment in terms of mercury accumulation in soils, plants, animals, or even human organisms is little recognized. However, mercury is present in small quantities in copper ore deposits, generally less than 1 g/Mg, but in a layer of copper-bearing shale (Polkowice deposits) its content was up to 1.89 g/Mg and in the Rudna extraction field up to 9.88 g/Mg [30]. Certain amounts of metalliferous dusts (containing mercury compounds) are emitted from copper smelters (Legnica, Głogów, and Orsk) and from Żelazny Most in Rudna, Poland – one of the largest in the tailing ponds in the world [31].

The aim of our study was to evaluate the total mercury accumulation in plants and farm animals in the area of copper industry influence, i.e., in the central region of Legnica-Głogów Copper District (LGOM).

Experimental

Study Area

We conducted our study in the LGOM area, which covers 14 villages of the Lubin, Polkowice, and Głogów districts. These areas are located near the Głogów and Cedynia copper smelters (CS); the Lubin, Polkowice-Sieroszowice, and Rudna copper mines/mining plants (MP); plus the ore enrichment plant (OEP) in Rudna. Mines, ore flotation plants, and smelters emit a lot of gaseous pollutants that contain metalliferous dusts (with mercury compounds). This is also the area where the tailing pond Żelazny Most and its wastes from copper ore mines is located. It covers an area of about 1,400 ha, and the volume of waste stored in it is ca. 500 mln m³. Mercury concentration of flotation tailings is 0.14-0.23 g/Mg, and in tank sludge supernatant it is 0.7 µg Hg·dm⁻³ on average [30-31]. Assuming an average Hg concentration in the waste of only 0.1 mg·kg⁻¹, its deposit in the disposal site would be about 50 Mg. Of course this is related to solid waste, but can also move to the liquid phase (aqueous) and then, through evaporation, get into the atmosphere. There are some data concerning total mercury content in the soil and drinking water of this region, confirming that the copper industry has impact on the local environment. Concentrations of Hg in soil samples ranged from 0.045 to 0.251 mg·kg⁻¹ [31] or even 0.57-4.64 mg·kg⁻¹ [32]. The permissible content of mercury in the soils from group A (protected areas) is 0.5 mg·kg⁻¹ DM, while in the soils from group B (at a depth of 0.3-15 cm) it is $3 \text{ mg} \cdot \text{kg}^{-1}$ DM, depending on water permeability [18]. Moreover, Barej et al. [33] reported that concentrations of total Hg in samples

Material and country	Content	Source*	
Air: Urban and industrial regions (different countries)	0.090-38 ng·m ⁻³	Kabata-Pendias and Szteke (2012)	
Contaminated soils (Poland)	Mean 1.5; max 4.6 mg·kg ⁻¹	Chojnacka et al. (2005)	
Contaminated soils (different countries)	0.12-17,000 mg·kg ⁻¹	Kabata-Pendias and Szteke (2012)	
Oceanic waters (North Atlantic and North Pacific)	Mean 0.20 ng·L ⁻¹	Driscol et al. (2013)	
Rivers water (Ghana)	<1.0-11 µg·L ⁻¹	Adjei-Kyereme et al. (2015)	
Stream sediments (Poland, Silesia)	<0.050-180 mg·kg ⁻¹	Pasieczna (2014)	
Coal (Poland)	Mean 0.074; max 0.32 mg·kg ⁻¹	Klojzy-Kaczmarczyk and Mazurek (2013)	
Coal ash (China)	0.13-4.8 mg·kg ⁻¹	Wang et al. (2006)	
Petroleum products (Poland)	4.7-15 μg·kg ⁻¹	Albińska et al. (2006)	
Petrochemical products (USA)	0.27-7100 μg·g ⁻¹	Wilhelm et al. (2000)	
Mineral-organic fertilizers (Poland)	0.0030-2.8 mg·kg ⁻¹	Górecki et al. (2002)	
Feed phosphates (Poland)	0.020-0.28 mg·kg ⁻¹	Hoffmann et al. (2004)	
Feed for poultry (Pakistan)	0.14-2.4 mg·kg ⁻¹	Imran et al. (2014)	
Feeding stuffs (EU: different countries)	Mean 0.030; max 1.3 mg·kg ⁻¹	Mercury as undesirable substances in animal feed, EFSA Journal 654, 1-76 (2008)	
Manure from farm animals (Austria)	0.080-0.58 mg·kg ⁻¹	Sager (2007)	
Compost: chicken manure + spent mushroom + soil (Spain)	0.29-0.86; max 17 mg·kg-1	Restrepo-Sánchez et al. (2015)	
Municipal wastewater (Italy)	Mean 1.4 mg·kg ⁻¹	Goi et al. (2006)	

Table 1. The content of mercury in environmental material according to different authors.

*available from authors

of drinking water collected at the LGOM region (seven farmsteads) ranged 0.3-4.4 μ g·dm⁻³, while the acceptable limit is on the level of 1.0 μ g·dm⁻³.

Except for the dominant role of the copper industry in this region (LGOM), there is also agriculture, i.e., highly productive cultivated soils or small-scale livestock farming; however, this demonstrates a downward trend [34].

Research Samples

The research material consisted of consumption and fodder plants (e.g., wheat, potatoes, and hay), as well as animal origin material such as milk, blood, and hair of dairy cows (breeding in small herds, grazing), eggs, and hen muscles and livers (outdoor breeding). The material was collected in the autumn from small-scale farms in Grodowiec, Grodziszcze, Gwizdanów, Komorniki, Krzydłowice, Tarnówek, Brodów, Mleczno, Orsk, Pieszkowice, Rudna, Rynarcice, Toszowice, and Żelazny Most. The samples were collected twice from 1-2 farms in the mentioned villages, i.e., in 2014 and 2015 (with cow hair in 2015 only). A total of 280 biological samples were analyzed (plant and animal origin material).

Plants were collected into plastic bags (wheat, hay, cleaned potatoes), while the milk was derived directly

from the cows (after milking). Blood samples for analysis were collected from the external jugular vein (*vena jugularis externa*), and the hair was cut out of the back part, then washed in water with detergent and rinsed in distilled water. Muscles (pectoral) and liver of laying hens (1-2 years old) were collected immediately after decapitation. Hen eggs were hard-boiled and egg albumen was mixed with egg yolk. Blood, milk, and potato samples were lyophilized. All concentrations of total mercury are given as µg·kg⁻¹wet weight (wet wt).

Similar material was not collected from other regions of the country (for comparative purposes), since mercury concentration in environmental and biological elements as well as in food is very differentiated, and it depends on numerous factors of natural and anthropogenic origin, which we demonstrated in our introduction and in Tables 1 and 2. Nevertheless, the results obtained in the two series of measurements were compared with the results of the studies carried out in this area (LGOM) in previous years (i.e., 2002 to 2007), and other studies by Polish and foreign authors.

Analytical Techniques

Analyses were performed using an atomic absorption spectrometer AMA 254 (Altec Ltd., Czech Republic)

Material and country	Content	Source*	
Cereals: various species (EU - 15)	0.0010-0.019 mg·kg ⁻¹	SCOOP 3.2.11 (2004)	
Infant cereals (Spain)	0.42-7.6 μg·kg ⁻¹	Hernández-Martínez and Navarro-Blasco (2013)	
Vegetables: various species (EU -15)	0.00040-0.011mg·kg ⁻¹	SCOOP 3.2.11 (2004)	
Potato tubers (Slovakia)	0.015-20 mg·kg ⁻¹	Klimakova and Poracova (2011)	
Edible mushrooms (Czech Republic)	0.25-13 mg·kg ⁻¹	Svoboda et al. (2006)	
Bee bodies (Poland)	0.50-6.4 max 23 μg· kg ⁻¹	Madras-Majewska and Jasiński (2005)	
Meat and fall (EU-15)	0. 0070-0. 012 mg·kg ⁻¹	SCOOP 3.2.11 (2004)	
Sea and river fish (Germany)	0.024-0.15. max 9.0 mg·kg ⁻¹	Schweinsberg (2003)	
Imported fish: tuna (New Zealand)	0.0070-0.66 mg·kg ⁻¹	Thomson and Lee (2009)	
Seafood (Chile)	0.070-1.9 mg·kg ⁻¹	Marko et al. (2014)	
Hen eggs, free range system (Poland)	mean 3.4 µg ·kg ⁻¹	Barej et al. (2015)	
Eggs of Canada geese (USA)	$4.6 \pm 0.32 \ \mu g \cdot g^{-1}$	Tsipoura et al. (2011)	
Cow milk (China)	1.0-3.9 µg·kg ⁻¹	Zhang et al. (2007)	
Women breast milk (Brazil)	<0.76-23 µg·L ⁻¹	Cunha et al. (2013)	
Human blood (Canada)	0.58-4.2 μg·L ⁻¹	Lambrinos (2014)	
Human hair (EU - 17 countries)	0.060-2.4 µg·g ⁻¹	Castano et al. (2015)	
Animal hair (Poland)	0.0040-0.018 mg·kg ⁻¹	Dobrzański et al. (2007)	
Hair of wild fur animal (Canada)	21-30 μg·g ⁻¹	Fortin et al. (2001)	
Food (human diet) USA	Mean 5.6 µg·kg ⁻¹	Iyengar et al. (2000)	

Table 2. The content of mercury in biological material and food according to different authors

*available from authors

to determine total mercury content directly in raw materials, regardless of Hg form. The limit of detection is 0.003 ng of Hg per sample. Several reference materials were used for verification of the method: soil (Loamy clay, No. RTC-CRM052-050), grass (Rye grass, No. ERM-CD281), and cow muscles (Bovine muscle, No. ERM-BB184) manufactured by IRMM (Belgium). The certified reference materials were analyzed with recoveries ranging from 97.5 to 104%.

Statistical Analyses

The results were analyzed statistically, the mean values and standard deviations for nine categories of plant and animal materials were calculated. The mean values between years (Series I and II) were compared using Student's t-test (computer software Statgraphics ver. 5.1) at the significance level of p<0.05.

Results and Discussion

Table 3 presents the results of plant origin materials examination. The maximum content of Hg in cereals (wheat) was 5.20, and the mean concentrations in particular series were 1.42 and 1.50 μ g·kg⁻¹. Thus, we noted a fourfold decrease of Hg content compared to previous results obtained almost 10 years ago [33]. Hg concentration in meadow grass (hay) was distinctly higher than in cereals. The maximum mercury content was 37.3 μ g·kg⁻¹, and the average values in particular series were 17.3 and 13.9 μ g·kg⁻¹, thus only a small difference in comparison to previous studies occurred [33]. The lowest concentration of mercury was found in potato tubers. The maximum content was 0.740 μ g·kg⁻¹, and mean concentrations in particular years were quite similar (about 0.300 μ g·kg⁻¹). It is difficult to compare these results, since previous studies relate to peeled tubers derived only in the area of Głogów Copper Mine (mean concentration of 0.0900 μ g·kg⁻¹) [28].

Polish authors reported a wide range of Hg concentration in plants, e.g., in grass from contaminated areas from 0.04 to 4 mg·kg⁻¹ DM, in cereals (grain) from different countries from 3.4 to 20 μ g·kg⁻¹ DM [3], and in potatoes 0.6 μ g·kg⁻¹ on average [35]. For comparison, the acceptable mercury content in basic feed materials (about 88% of dry matter) is 0.1 mg·kg⁻¹ DM [36]. According to EFSA data [37], Hg content in complete feed mixture for terrestrial animal categories in the EU is 0.012-0.053 μ g·kg⁻¹, on average. The EU legal act

Series	Unit	Grain	Meadow grass	Potato
Ι	Mean	1.42	17.3	0.350
	SD	1.70	7.30	0.140
	Range	0.200-5.20	7.50-37.3	0.160-0.700
II	Mean	1.50	13.9	0.270
	SD	0.910	5.72	0.180
	Range	0.100-3.80	8.90-33.6	0.400-0.740
Earlier study	Average	5.14*	18.5*	0.0900 **
	Maximum	6.67*	45.0*	0.700**

Table 3. Total mercury content in plant materials in $\mu g \cdot k g^{-1}$ wet wt.

*acc. Barej et al. (2009); ** acc. Wyka et al. (2009)

concerning chemical contaminants in food does not include mercury, except in fish and fish products [38].

Mercury concentrations noted in animal origin products were generally lower than in plants (Tables 4 and 5). Maximum Hg content in raw milk was 0.400 μ g·kg⁻¹, with similar means in both series (0.230 and 0.200 μ g·kg⁻¹). In previous studies [33], mean content was 8-fold higher. Maximum concentration in the blood of cows was 6.00 μ g·kg⁻¹, with similar means in both series (about 1.80 μ g·kg⁻¹). Previous studies conducted in other industrialized regions of Lower Silesia (near Wrocław) demonstrated an average Hg concentration in blood of cows on the level of 0.950 μ g·kg⁻¹, while in their hair it was 2.49 μ g·kg⁻¹ (maximum 6.20 μ g·kg⁻¹) [39].

Low mercury concentration in milk samples (7-8 times lower than in blood) should be emphasized when evaluating the results of these analyses (cattle), while it shows the protective function of the mammary gland. There is no current legislation that sets acceptable limits of mercury content in milk, but, for example, average values of Hg in cow's milk (and milk products) in EU countries at the turn of 20th and 21st centuries ranged from 0.5 to 5 µg·kg⁻¹ [40]. Another good example of an environmental pollution bioindicator is toxic metals concentration in animal hair. The values obtained are relatively lower compared to other domestic animals [3, 41-42], but similar to the results of previous studies [39]. It is interesting that mercury content in horse hair is positively and significantly correlated with other investigated elements, such as arsenic, silicon, phosphorus, sulfur, and selenium [43].

Maximum Hg concentration in egg content was $3.50 \ \mu g \cdot kg^{-1}$, while average values in two subsequent series were on the level of 1.54 and 1.13 $\ \mu g \cdot kg^{-1}$. Determined Hg concentration were 2-3 fold lower compared to previous studies of samples from this area [33]. Twice lower concentrations of Hg were found in the muscles of hens, and the maximum content was 1.80 $\ \mu g \cdot kg^{-1}$. Compared to previous studies, determined concentrations were multiple lower, taking into account that the average value of Hg detected in earlier years was 6.00 $\ \mu g \cdot kg^{-1}$ [44].

Series	Unit	Milk	Blood	Hair
Ι	Mean	0.230	1.86	ns
	SD	0.120	1.51	-
	Range	0.100-0.400	1.00-6.00	-
II	Mean	0.200	1.77	2.88
	SD	0.130	0.360	1.16
	Range	0.100-0.400	1.10-2.20	0.100-6.20
Earlier study	Average	1.65*	0.950**	2.49**
	Maximum	3.50*	2.00**	6.52**

*acc. Barej et al.(2009); **acc. Dobrzański et al.(2009); nsno study

Table 5. Total mercury content in animal materials (hen) in $\mu g \cdot k g^{-1}$ wet wt.

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Series	Unit	Egg	Muscle	Liver
	Mean	1.54	0.660	2.63
Ι	SD	0.950	0.510	3.91
	Range	0.300-3.50	0.100-1.80	0.400-16.0
II	Mean	1.13	0.700	2.27
	SD	0.670	0.450	1.24
	Range	0.200-1.90	0.100-1.60	1.10-5.30
Earlier study	Average	3.86*	6.00**	7.10**
	Maximum	6.90*	11.0**	18.0**

*acc. Barej et al. (2009), ** acc. Kołacz et al. (2003)

However, an average Hg content in hen's liver samples was within a wide range of 0.400 and 16.0 μ g·kg⁻¹, but the average values in both series were quite similar (between 2.27 and 2.63 μ g·kg⁻¹). Mercury concentrations in previous studies were almost three-fold higher [44].

There is still a lack of literature in the field concerning mercury content in poultry products. An average Hg content in eggs in the EU countries at the turn of 20th and 21st centuries ranged from 1.3 to 2.0 µg·kg⁻¹ [40], although the level of 0.5-4.0 μ g·kg⁻¹ was observed in backyard farming in Belgium [45]. The concentration of mercury in poultry tissues and organs depends on the housing system as well as on the quality of feed and water. For example, an average of 7.06 mg·kg⁻¹ of mercury was found in drinking water at a broiler farm in Saudi Arabia, which resulted in increased Hg concentration in liver samples of chickens up to a level of 1,167 μ g·kg⁻¹ [46]. Mercury also easily accumulates in egg content, liver, and muscles when methylmercury is presented in the feed [26]. This can be prevented to a limited extent when feed additives containing humic compounds and aluminosilicates are supplemented [26, 47]. Another solution is selenium supplementation. Se is an antagonistic element to Hg, but it can easily be overdosed and then is highly toxic in excess [1-3].

According to EFSA recommendations [22], the levels of Hg present in foods (other than fish and seafood) are of lower concern mainly because the forms of Hg present in these other foods are not methylmercury. Thus the maximum levels for Hg in foodstuffs were set only for fishery products and muscle meat of fish (0.5-1.0 mg·kg⁻¹)[38]. Taking into consideration that the highest concentration of Hg determined for the present study were on the level of 37.3 μ g·kg⁻¹ (meadow grass) and 16.0 μ g·kg⁻¹ (laying hens liver), which is much lower than the acceptable content in fishery products, there is no possible health risk (mercury criterion) for animals and humans who consume food products from the investigated area.

Conclusions

The obtained results indicate a low level of total mercury pollution in an area of copper industry influence. The highest concentrations of Hg were found in grass and hen's liver samples. The lowest concentration of Hg was noted in cow's milk and chicken muscles ($<1 \ \mu g \cdot kg^{-1}$). There was a many-fold decrease of mercury concentration in cereals, cow's milk, and poultry products (eggs, muscles, liver) in comparison to previous studies (2002-07), which indirectly proves the effective environmental efforts led by local authorities and industry.

The investigated copper industry does not pose a toxicological concern (mercury criterion) to the natural and agricultural environment in the area of its influence. However, periodic biomonitoring studies are necessary, considering physico-chemical and biological effects of this highly toxic element, as well as the EU guidelines concerning the restrictions in Hg emissions.

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