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Protecting Fish Consumers Against Excessive Exposure to Mercury

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Article

Arithmetic vs Weighed Means in Fish Fillets Mercury Analyses

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Abstract: Mercury (Hg) analyses in species of fish are performed for two reasons 1) to safeguard human health, 2) to assess environmental quality, since different environmental changes may increase the Hg concentrations in fish. These analyses are important since both natural and human activities can increase these Hg concentrations, which can vary extensively, depending on the species, age and catching location. Hg contaminated fish or other marine foodstuffs can be only detected by chemical analysis. If the aim of Hg analysis is to protect the health of marine-food consumers, researcher workers must consider the location where the fish were caught and interpret the results accordingly. Health and environmental officials must appreciate that in specific places local people may have a daily diet entirely consisting of fish or other marine foods, and these individuals should not be exposed to high concentrations of Hg. Regional and national health and environmental officials should follow the recent guidance of international organizations when drawing their final conclusions about whether the products are safe or unsafe to eat. Correct statistical calculations are not always done so too high Hg-amounts could be presented, and the fish eaters could be protected. This work has been done to show the differences in Hg concentrations between weighed (weighed with fish weights) and arithmetic means. Thus the only weighed mean also includes the Hg contents in fishes so that the exposing of Hg can be evaluated.

Keywords: food contamination; health protection; methyl mercury; sulphate reduction; weighed mean

1. Introduction

The Hg analyses in fish starts from selection the fishing areas where there are suspicions about the Hg concentrations and possibly other contaminations in the marine environment, which will then be fished. The fresh or frozen fishes are then taken to the chemical analyses most often undertaken by highly trained personnel using expensive equipment [1] with the analyses typically being performed in a few accredited laboratories [2] at high costs. The last part involves the interpretation of the values with a valid statistical evaluation to allow the drawing of reliable conclusions whether fishes are safe or not safe.

Mercury toxicity became evident after some 80 years use of Hg compounds as medical compounds and as horticultural and agricultural pesticides [3,4]. The Minamata (Japan) tragedy was an alarm call, since Hg compounds released from a battery-producing factory had heavily contaminated the fish and other marine organisms in Minamata Bay in the south-western coast of Japan [5]. It caused a serious nervous disease in hundreds of people and even some deaths. Thus, in 1959, the Japanese Ministry of Health and many international and national organizations started efforts to ensure that this disaster could never again happen. The first restrictions and prohibitions were extended to different Hg compounds [6,7]. For example, the World Health Organization (WHO), the Food and Agriculture Organization (FAO) set limits for how much Hg could be present in fish intended for human consumption [8]. It was appreciated that both marine and freshwater fishes and other products from the sea were the most critical Hg sources in the human diet. It was evident that the muscles of some large predatory fish tended to contain more Hg than those fish

which were herbivores. Two other factors found to be important were the industrial contamination of a fishing site and the size of fish to be consumed [8].

Different organizations started to develop analytical methods to detect both inorganic Hg and methyl Hg. After the Minamata tragedy, the levels of Hg compounds in fish and other marine food have been monitored especially near industrial enterprises, because the too high mercury concentrations in different foods can be a serious obstacle for marketing these products. Therefore, also commercial enterprises became concerned about Hg and started to analyze its concentrations in their own specific products.

Today, the European Union (EU) has set two regulations, 2022/617 and 2023/915 [9,10], concerning the analysis of Hg in food products. The present regulations allow that fresh weight of fillets of the most edible fish species can contain a maximum of 0.5 mg/kg total Hg but for some very commonly used fish products such as sardines, anchovies and herrings, the limit is lower, only 0.3 mg/kg Hg. In contrast, some predatory fishes can contain a higher level of 1.0 mg/kg Hg. Today, in many countries the recommendation is that humans should only seldom (or in some cases, never) consume those fish species which can contain the highest concentrations of Hg.

1.1. Sulphate Reduction and Methyl Mercury Formation

The formation of methyl mercury has been known for a long time, e.g., in the 1990s the first publications described that organic methyl mercury compounds could be formed during sulphate reduction [11,12]. Many anaerobic sulphate reducing bacteria possess an ability to produce methyl mercury as a side reaction from inorganic Hg compounds [12]. This reaction happens in freshwaters, marine waters, and wetlands both in tropical and arctic climates. The reaction can occur if inorganic Hg and sulphate are present at a low redox potential. The layers between water and sediments are thus good places for methyl Hg formation [12,13].

Sulphate-rich wastewaters accumulate in bottoms of waterbodies because their specific weight is higher than that in surface waters with low sulphate concentrations. Thus, the sulphate concentration in the bottom layers of waterbodies and for example, under stones or in gravel and sand will be the locations with the highest sulphate concentrations. An increasing concentration of sulphate decreases the redox potential and increases the risk of a greater reduction of inorganic Hg into methyl mercuric compounds [12,13]. It is evident that global mercury deposition is sufficient to allow Hg methylation. It is known that the Hg can originate from volcanic eruptions, or from human activities such as coal burning, gold mining using elemental Hg etc. Similarly, the level of sulphate needed to allow sulphate reduction and methyl mercury formation can be as low as 20 – 40 mg/L sulphate, and this level can originate from industrial emissions or from fertilizers [13,14]. Unfortunately, the lake water just above the sediments are seldom studied and at least in Finland they do not belong to obligatory analytical sites in environmental monitoring.

1.2. Toxicity of Methyl Hg

The formed methyl Hg compounds are more toxic to humans than the corresponding inorganic compounds [15] and the exposure dose of methyl Hg must always be considered. Methylated Hg compounds have a clear tendency to bioconcentrate via routes: bacteria, other microflora, small fishes, larger fish and mammals (including humans) and birds. Bioconcentration happens because the methyl Hg compounds are more fat-soluble than inorganic mercury compounds and thus, they remain stored in fat tissues and are not available for metabolism and excretion. Therefore, the European Union Food Safety Authority [15] has issued (and updated) its scientific opinion about methyl mercury.

Three major messages of this scientific opinion report [15] are:

1. As much as 90 % of total Hg in fish can be present as methyl mercury.
2. Small children and pregnant or lactating women and even women who potentially can become pregnant belong to the most sensitive individuals and they should avoid eating fish rich with methyl mercury.

3. The safe weekly dose of methyl mercury for one person is proposed to be only 1.3 µg/body weight (kg).

The protection of the pregnant women is important since Hg compounds are teratogenic. Furthermore, with respect to infants, the weekly Hg limitation means that a small child with a body weight of 10 kg could consume only 0.013 mg (13 µg) methyl Hg, i.e., a child should eat no more than 29 g of fish per week, if one assumes that the fish would contain 0.5 mg/kg total Hg of which would be 0.45 mg/kg methyl mercury. In practice, it means that families with small children and /or pregnant women must be confident that the fish on their dinner table should contain only low amounts of Hg and the fish should originate from an area known to be safe. Furthermore, the family should not eat fish too often even if the children themselves have participated in the catch.

The protection of fish consumers is possible if Hg and methyl Hg analyses will be achieved, and the amounts of Hg are known. The major aim of this paper is to show which type of mean counting method should be used to evaluate the true amount of Hg and health risk of Hg concentrations of perch intended for human consumption. It should be possible to analyze the total Hg amounts, which the fish eater can be exposed, and methyl Hg can be estimated to be 90 % of the total Hg [15]. The trustful results could also help fishers who sell a part of their catch to companies, since these fish need to fulfill all legal requirements (including the Hg-concentration), which may be analyzed in foreign customs laboratories. The major problem is to use an appropriate way to determine if the edible fish contains too elevated levels of Hg so that the fish-eaters will be protected.

2. Materials and Methods

2.1. Fish Analyzes

The Water Framework Directive of European Union [16] states that all European waters should have at least a good ecological status and thus the fish and the other living organisms should be acceptable for human consumption. However, it is evident that the Hg-concentration in fish depends on their living area, position in the food chain, size, and age [17,18]. Thus, the Hg concentration in fishes can vary extensively – but mainly it varies according to the size and age (weight and length) of the fish.

The analyses of mercury should be conducted in an accredited laboratory which should adhere to the protocols listed in reference texts [9,10]. The analytical values presented here were performed by the Eurofins Environmental Testing Finland Oy, which is an accredited laboratory participating in quality control analyses with the same specimens being assayed in many laboratories in different countries. There are several stages to the evaluation of Hg concentrations in fish e.g., the dissection of the sample to obtain fillet specimens, followed by Hg extraction with the final analysis performed by gas chromatography coupled to mass spectrometry. There is always a degree of uncertainty associated with each Hg determination (as is the case in all chemical determinations) and the uncertainty found for some Hg assays can be as high as $\pm 25\%$. The EU Commission's regulation [19] offers some assistance about this problem although it should be included in the recommendations.

2.2. Chemical Analyses and Data Counting for Hg

The fishes were frozen and kept frozen until Hg analyses so that volatile Hg compounds would be preserved. The determination of metals in fishes included Hg and some other metals (these concentrations were low, and this data is not presented in this work; furthermore there are no limits set by European regulations) [15]. The analytical values presented here were performed by the Eurofins Environmental Testing Finland Oy, which is an accredited laboratory.

In mathematics, the mean of Hg concentration can be calculated in three ways; 1) as a simple arithmetic mean (average) or 2) as a geometric mean or 3) as a weighted mean (according to the fish's weight). The arithmetic mean counts only the mean of Hg concentrations in different fishes. Geometric mean counts the mean after logarithmic formation. Arithmetic or geometric mean do not consider the amount of Hg so that they do not warn of too high exposure Hg concentrations.

The weighing mean counts first the sum of Hg amounts in all fishes and this sum of Hg amounts is then divided with the sum of fish all weights. The next equation shows the detailed counting way for Hg amounts in fishes.

$$X = \sum w \cdot c / \sum w$$

where X is the weighted mean and $\sum w \cdot c$ is the sum of weights for all fishes multiplied with the corresponding Hg concentrations and $\sum w$ is the sum of weights of all fishes. Thus $\sum w \cdot c$ also presents the total amount of Hg in all fishes.

The international regulations do not describe the exact detailed ways to calculate final results [9,10]. These recommendations do not inform the health inspectors what should be done if many but not all the analyzed fishes contain more than the limit value of Hg or if most of edible fish fillets do contain more than the limit value, but nonetheless the arithmetic means are less than the limit value. This phenomenon can happen especially if there are large variations in the size of the fishes i.e., there are many small-sized fishes which contain only low concentrations of Hg but there are some large ones which contain high concentrations of Hg.

Besides fish analyses the same Eurofins laboratory also analyzed lake waters from surface, middle and from 1 m bottom using standard methods. These analyses included sulphate and other physico-chemical parameters, but only sulphate concentrations have been discussed here.

2.3. The Lakes

The selected lakes: Laakajärvi and Kiltua are situated in the southern side of the Talvivaara mining area (today belonging to Terrafame company) in the eastern part of central Finland (app. 63°50'N, 27°50'E). Both these lakes are situated outside of the mining area, so that the fish should be assumed to be safe for human food consumption, and the mining company has no permission to contaminate these lakes via its emissions. Lake Laakajärvi is the first waterbody downstream after the industrial mixing zone of the Terrafame mining area. Lake Kiltua lies downstream of Lake Laakajärvi; there are short rivers connecting Laakajärvi and the mining area's water reservoir and also between Lake Laakajärvi and Lake Kiltua. In Lakes Laakajärvi and Kiltua, the levels of Hg and other values in fishes and water are set by the Water Framework Directive [16]. The area of Lake Laakajärvi covers 34.7 km²; Lake Kiltua is smaller, 10.1 km².

2.4. The Used Report

The fishing for this work was done in autumn 2014 for the Talvivaara company (today mining is belonging to Terrafame) as a part of its obligatory environmental control. According to Finnish Environmental Law [20], this data must be made available for all people living in the mine's vicinity including fishers and fish-consumers, who require this information about the quality of the fish catch and other environmental issues. The report used in this work originates from 2014 [21], since this is the last published report which also included data about all individual fishes with their sizes and Hg-concentrations. The report [21] was paid, edited, and finally approved by Terrafame or Talvivaara company before its release to the local environmental officials, who then were tasked with either accepting or rejecting the report's findings. This report made it possible to calculate the arithmetic, the geometric and the weighted means (weighted according to each individual fish's weight). The newer reports have presented only arithmetic means.

The only fish species selected for this work is perch (*Perca fluviatilis*) since the number of northern pikes (*Esox lucius*) was often too low. Nonetheless, perch is a suitable choice, since it is a popular fish with a good taste, and it can be easily fished even by children and families for their own consumption. One major problem with the very small perches with very small fillets is that it is laborious to obtain pure fillets after removal of the fish's scales, bones, and gut, without the spillage of bile.

The results of Table 2 have been calculated with the Microsoft Excel program.

3. The Results

The results about the individual perch sizes and Hg concentrations are shown in Table 1. The original data is published only in Finnish [21]. The author has set the data in a new order so that the results may be easier to discern, and the four separate tables listed in [21] have been merged into Table 1.

Table 1. The weights and lengths of the perch and their total mercury (Hg) concentration of the edible part (fillets) in two lakes [21].

Fish weights (g)	Lengths (cm)	Total Hg concentration in fillet (mg/kg)
Lake Laakajärvi		
41	16.7	0.28
41	16.2	0.20
42	16.0	0.18
63	18.3	0.23
74	19.0	0.26
288	28.7	0.74
333	31.0	0.85
392	30.0	0.60
436	32.4	0.89
546	34.0	0.80
Lake Kiltua		
103	31.5	0.34
122	22.3	0.39
144	22.8	0.38
144	24.5	0.52
156	24.1	0.53
173	25.4	0.67
175	25.4	0.73
190	22.7	0.72
256	24.0	0.57
265	24.9	0.73

Table 2. Different means of the Hg-concentrations in the ten perches studied, and the fishes divided by the weights of the fish to estimate those exceeding or not exceeding the Hg-value of 0.5 mg/kg and their numbers and total amount of Hg in all ten perch samples as well as the correlation coefficients between the amounts of Hg and the weights of the fish and correspondingly between the amounts of Hg and fish lengths in both Lakes Laakajärvi and Kiltua.

	Lake Laakajärvi	Lake Kiltua
Arithmetic mean ± standard deviation (mg/kg)	0.50 ±0.30	0.55±0.15
Geometric mean (mg/kg)	0.42	0.53
Weighted mean (weighted according to fish weight) (mg/kg)	0.72	0.59
Total weight of all ten perch (kg)	2.254	1.728
Total weight of perch containing less than 0.5 mg/kg Hg (+ their number) (kg)	0.261 (5)	0.369 (3)
Total weight of perch containing more than 0.5 mg/kg Hg (+ their number) (kg)	1.995 (5)	1.359 (7)
Total amount of Hg in all ten fish (mg)	1.62	1.01
Correlation coefficients between weight and Hg-concentration	0.92	0.70
Correlation coefficients between length and Hg-concentration	0.91	0.92

The sulphate concentrations in water during some previous years are presented in another report [22]. The concentration of sulphate was 628 mg/L in the bottom of Lake Laakajärvi in the autumn of 2014. The sulphate concentration in the bottom of Lake Kiltua was 37 mg/L in autumn 2014.

4. Discussion

4.1. The Different Means

The arithmetic means and geometric means for Hg are near to 0.5 mg/kg. However, the arithmetic or geometric means do not give any realistic information about the actual amounts of Hg that the fish consumers were exposed, if some fish contain only trace amounts of Hg, and others, the large fishes had very high concentrations. It is impossible to estimate the exposure of Hg from either the arithmetic or geometric mean, which is crucial when one is estimating human exposure [15].

It should be noted, however, that the standard deviation especially that in Lake Laakajärvi was remarkably high indicating that the fishes varied extensively in several parameters. As is evident from Table 1 where there were large variations in both the sizes and the Hg-concentration in the perch, and all larger fishes contained more Hg than their smaller counterparts, a fact that has been reported internationally previously [15]. In addition, the correlation coefficients between the length of the fish and their Hg contents and between weight and Hg contents are high, again in accordance with previous publications [8,11,13,14].

The weighted means are the highest of all means (Table 2), since all the larger perches, from which good-sized fillets could be made, also contained the highest amounts of Hg. Thus, in Lake Laakajärvi, the smallest five perches (all with Hg-concentration less than 0.5 mg/kg) represented less than 12 % of total perch fillet mass, and correspondingly almost 89 % of perch mass in the five larger fishes contained more than 0.5 mg/kg Hg (Table 2).

In Lake Kiltua, the three smallest perches accounted for some 21 % of perch mass while the percentage of the perch mass which contained over 0.5 mg/kg Hg, was almost 79 % but in two of these specimens, the level of Hg was only slightly above the proscribed limit and their values may not be considered due to methodological uncertainties. In fact, in both lakes, the fish in this “larger” category were not particularly big with respect to how large fish in this species can be, nonetheless their Hg concentration did exceed the proscribed limit, even considered methodological uncertainties.

It is often estimated that some 40 % of fish weight is the edible part in fillets [23]. In Lake Laakajärvi, all perches taken for this work had a common weight of almost 2.2 kg (Table 2), and thus these fishes could be turned into about 880 g of perch fillets for food. This fillet mass could be enough for one substantial meal for five or six persons if a portion is estimated as 150 g as presented in [23]. The consumers would thus be exposed to around 1620 µg of total Hg and around 1460 µg methyl mercury, if 90 % of Hg would be in the methyl mercury form [15]. If this would have been their only weekly fish dinner, then the only way that their weekly exposure dose of methyl mercury could be limited to 1.3 µg per kg of body weight [15], would be that the total weight of those six people eating the perch should be 1120 kg, which is most unlikely.

Both the total Hg and methyl mercury levels were lower (1010 µg and 909 µg) in Lake Kiltua perches since the total weight of these ten perches was 1.73 kg giving around 700 g in fish fillet form. By using the same calculation, five people could consume these fillets. If that were their only fish dinner for a week, in order that they would not be exposed to an excess of Hg (and its methyl Hg form), and their total body weights would also need to be heavy – almost 700 kg.

In both these cases the safe amount of perch fillet that should be served would be much smaller than what is commonly consumed by these lakeside inhabitants (150 g). In both these lakes, the mass of perch fillet on the plates should be reduced to less than 100 g per dinner.

If one critically examines the different means and their ability to protect the health of fish consumers against Hg risks, it is difficult to see any reason for utilizing the arithmetic mean, since the arithmetic mean does not consider the size of the fish and thus the Hg content of the portion of fish on the dinner plate. Only the weighted mean provides an indication of the total Hg exposure; this is clearly mentioned in an EU report [15] and in the Codex Alimentarius guidelines of FAO and WHO [24]. Both these international reports, made by groups of international experts, strongly emphasize the need to give details of both the total exposure and the maximum weekly intake of Hg

(or other pollutants). In fact, the weighted mean has also been used in several scientific publications which have evaluated the exposure by diverse human groups to different pollutants [25–28]. The concept of the weighted mean is also applied when considering the different nutrients in human food or animal feed such as the consumption of proteins, carbohydrates, fatty acids, and different vitamins.

Thus, details of the weighted mean of Hg would protect the fish-eating population better than the arithmetic mean, or geometric mean despite the fact it gives the highest Hg concentrations. The weighted mean takes more into account the contribution of larger fishes, as these will make up the greatest amounts of actual fish on the plate, if the entire catch would be consumed.

The result is not very trustful if the weight of perches (in this case) varies from 41 g to 540 g so that the Hg-concentrations also vary highly, and the numbers of pikes was so low that they should be omitted. The companies who must present fish results, must take enough time to make a careful fishing so that the numbers of fishes are at least ten (if that is the claim). In addition, the size of all fishes of the same species must be moderate and typical for those fishes which will be consumed.

4.2. Why the Fishes in These Lakes Contained too Much Mercury

We can speculate about the reasons for the excessively high Hg levels in fish in these lakes. The general atmospheric fallout of Hg is evident Hg source also in this part of Finland [11,12]. The sulphate concentrations were high at the bottoms of the both lakes in the report of the 3rd annual cycles [22]. The sulphate concentration of 628 mg/L is very high comparison of the maximal sulphate concentration of 12 mg/L SO_4 in 36 Finnish forest streams monitored for 20 years [29]. Evidently the high sulphate concentration in the bottom of lake waters is a sign that there was unsuccessful complete mixing of the different layers of the lake's water layers i.e., oxygen in the surface layers was not being transported to the bottom (typically these lakes are dimictic with two annual complete mixings -in spring and in autumn due to temperature differences in the water layers). Due to disturbance in the full water mixings, there have been high sulphate concentrations in the bottom of Lake Laakajärvi and the most sulphate-rich water with the highest specific weight has survived in bottom of Lake Laakajärvi allowing methylation of Hg and caused high Hg concentration in fishes.

In Lake Kiltua, the highest sulphate concentration in the bottom layer of water was 37 mg/L which may indicate that complete mixings of the waters may have occurred in spring and/or autumn and therefore the methylation of Hg was lower.

A massive accidental leakage from Talvivaara mining area occurred in 2012 between November 4th and Nov 15th when at least 200 000 m³ of highly contaminated gypsum precipitation solution spread into the natural waters and forests towards Lakes Laakajärvi and Kiltua. During this event, high amounts of acids were formed i.e., the pH measured in the leaking wastewater was at pH 2.89 instead of the aimed pH 9 and this polluted water contained high concentrations of heavy metals and sulphate [30]. It is likely that sulphate-rich wastewater continued to reach Lakes Laakajärvi and Kiltua.

4.3. The Local People and Animals as Fish Eaters

Many farmhouses, other permanent dwellings and summer cottages are situated along the shores of Lakes Laakajärvi and Kiltua. Some families have lived in this area for generations and or these people, a fish diet was their staple. Fish represents still the major source of Hg throughout Finland [31]. In addition, while many local fishers may sell a part of their catch, others may eat the fresh fish from their home lake many times a week, especially if they catch a large fish. It is evident from the published report [21] that there were no truly large perch although specimens as large as 1 kg can be caught, and as stated, it is the largest fish which contain the highest concentrations of Hg. In addition, the release of more relevant and reliable information could have improved the image of the mining company, and it would have had more confidence that the local fishes are safe for human consumption, especially among the people living in its vicinity. If the mining company can demonstrate that to be the case, it could also have increased the social acceptance of mining activity in this area. In addition, many wild waterbirds and wild animals are adept at catching fish (e.g.,

perch) i.e., the perch is an important part of the natural food chain. In principle, the environmental norm value for Hg has been set at 0.25 mg/kg for perch to protect fish-eating birds and some mammals [32]. Obviously, the lower the concentration of Hg and organic mercury in the environment protects better the wildlife.

Mining producing may be important in future despite water contaminations by Hg and other toxic compounds. New methods for detecting methyl Hg [33–35] are welcome despite they are still in development phase.

5. Conclusions

The **weighted** arithmetic mean (weighted according to fish weight) is a better way of informing consumers of fish about its safety. This type of calculation is more informative than other ways to calculate averages. In addition, it is the larger fish that can be the decisive factor in determining the actual Hg exposure in humans. Therefore, EFSA and the other international institutes should recommend that in the future national health inspectors should use weighted means when examining the concentrations of pollutants in fish. The local health officers must protect humans from being exposed to high levels of Hg and in this way, they can promote the activity of local fishermen to sell their catch both locally and even internationally as it will be safe for human consumption.

The health officers should be familiar with many statistical tests. They need to be taught when, why and how to calculate the weighted means when estimating the human exposure to Hg in fish. Therefore, they should receive detailed guidance on how this calculation should be done. The best way is to give examples like those listed in Tables 1 and 2. The local health and environmental officers should be able to follow the scientific development so that they know the reports of EU or FAO & WHO [15,27]. In the future, the environmental impact reports as that [21] should include all individual Hg results as well as the linked weights of the fish such as was provided in [21] so that health and environmental research officials can also count the weighted means.

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