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EXPOSURE OF THE POPULATION OF THE REPUBLIC OF MOLDOVA TO MERCURY AND THE WAYS OF REDUCING HEALTH RISK

331.02 HYGIENE

Summary of the thesis of Doctor of Medical Sciences

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The thesis was developed in the Scientific Laboratory of Chemical Hazards and Toxicology of the National Agency for Public Health

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LIST OF ABBREVIATIONS

Hg ^{Sv} _{PCM}	Mercury screening value in fish, crustaceans and molluscs					
b.w.	body weight					
CV	Variation coefficient					
DcQ	Daily consumed quantity					
DHA	Docosahexaenoic acid (omega-3 fatty acid)					
DI [Hg]	Daily intake of metallic mercury					
DI [MeHg]	Daily intake of methyl mercury					
Dn	Own data – with reference to mercury concentrations in fish, crustaceans and					
որ	molluscs					
DrQ	Daily required quantity					
EFSA	European Food Safety Authority					
EMEP	European Monitoring Emission Program					
EPA	Eicosapentaenoic acid (omega-3 fatty acid)					
EU	European Union					
FAO	Food and Agriculture Organization of the United Nations					
FCM	Fish, crustaceans and molluscs					
FOREGS	Forum of European Geological Surveys, Geochemical Baseline Programme					

CEMS/EOOD	Global Environment Monitoring System - Food Contamination Monitoring							
GENIS/FUUD	and Assessment Programme							
HI	Hazard Index							
HS	Harmonized Commodity Description and Coding Systems 2017							
IDB	International Databases - with reference to mercury concentrations in fish,							
	crustaceans and molluscs							
IES	State Ecological Inspectorate							
IN	National Inventory of Mercury Emissions of the Republic of Moldova 2014							
ITC	International Trade Centre							
LOD	Limit of detection							
LOQ	Limit of quantification							
MC	Minamata Convention							
MeHg	Methyl mercury							
ML	Maximum Limit							
MSC-E	Meteorological Synthesizing Centre - East							
NAPH	National Agency for Public Health							
NBS	National Bureau of Statistics							
Omega – 3	Omega -3 long chain polyunsaturated fatty acids (DHA + EPA)							
RfC	Reference Concentration							
RfD	Reference Dose							
RM	Republic of Moldova							
R _t C _a	Theoretical allowed consumption rate							
UNEP	United Nations Environmental Programme							
WHO	World Health Organization							

THE RESEARCH CONCEPTUAL FRAMEWORK

Research actuality and importance. According to the World Health Organization (WHO), exposure to mercury (Hg) even in very small amounts can cause severe effects on the population health or to foetus due to exposure of their mother's during their pregnancy. Mercury affects the nervous, digestive, cardiovascular and immune systems [20, 42]. In the National Profile on sustainable management of chemicals in the Republic of Moldova (RM) from 2008 [2], Hg is set as a grade 1 priority at the national level, statement reconfirmed by a subsequent national report in 2012 [31]. The RM bears coherent and complex statistical data regarding Hg assessment or as a factor of the environmental pollution and population's health impact [34, 35, 36, 37, 38, 45].

Between 2014-2017, the main sources of Hg pollution in the RM were identified. The Environmental Pollution Prevention Office under the Ministry of Environment has prepared the National inventory on mercury releases in the Republic of Moldova (IN) for the reference year 2014, the first national report in this regard. Was established that there are currently sources of Hg pollution in the country, being identified emissions of 972,12 kg Hg/year [33].

The RM has ratified the Minamata Convention (MC) recognizing that Hg is a chemical with worrying effects worldwide due to its long-distance transport capacity, persistence in the environment, bioaccumulation capacity in ecosystems and significant negative on human and environmental health [24]. At the same time, authorities become aware of the risks on vulnerable population's health - women, children [26, 30, 42, 44]. Thus, the health authorities by articles 16 and 19 of the MC are obliged to inform, raise awareness and educate the population as well as to research, and to monitor the levels of Hg in the environmental elements, food products with the assessment of the impact on the population's health [24, 32, 37, 45].

For these reasons, the performed study is currently of a major importance for the public health policy, specifying the vital relevance of knowing the level of Hg in water, air, soil and food, especially in marine products, and setting the necessary recommendations.

The study purpose: Mercury exposure assessment for the population of the Republic of Moldova for the elaboration of prevention measures appropriate to the appraised degree of risk.

Objectives:

- 1. Identification of potential sources of mercury pollution.
- 2. Determination and analysis of mercury concentration in frequently consumed species of fish, crustaceans and molluscs (FCM).
- 3. Estimation and analysis of mercury concentration in air, water and soil.
- 4. Risk assessment for the exposure to mercury of the population and development of prevention measures appropriate to the degree of appraised risk.

General methodology of scientific research. An ecological study was carried out using complementary descriptive, calculus and experimental methods to assess and estimate the impact of mercury (metallic form, methyl mercury) on the population health of the RM in compliance with scientific rigors, ethical principles of institutional, national and international research. The study object was considered general population and children from boarding schools aged 7-18y, kindergartens with 9,5-10h activity regime assuming that the respective category is aged 3-7y and children from nurseries with activity regime 9,5–10h, aged 1-3y. Daily intake for methyl - mercury and metallic mercury was calculated. We applied a toxicokinetic model to calculate the theoretical MeHg concentration in hair and blood. Using analytical laboratory tests the Hg concentration in various fish species, crustaceans and molluscs, in air, water and soil was investigated.

The novelty and scientific originality of the results obtained. For the first time, from a systemic perspective, was performed a complex multilateral study of the level of Hg pollution of the environment, toxico-hygienic aspects on the Estimated Daily Intake of Hg, metallic and organic form, by human body for the general population and children of the Republic of Moldova (RM) by applying different approaches. It has been scientifically argued which necessary measures should be taken by the public health service. The nationally recommended fish consumption (Daily Required Quantity) for children of different ages was analysed relating to the presence of Hg in FCM. The study was conducted by applying a new, justified, innovative methodology for the RM.

The exposure level to Hg was identified for the RM population through water, air, soil, food (fish, crustaceans and molluscs) and the intentional use of Hg in devices. The peculiarities of the exposure to Hg for the general population (per capita) and children of different ages in the RM were calculated, characterized, analysed and scientifically proved for better understanding of the problem size at national level. The study aids to the development and optimization of public health strategies and prevention measures aimed at mitigating the impact of Hg on the public health.

The research results magnify the theoretical basis part of estimated population's health associated risk as a result of exposure to Hg by ingestion of soil, water, air and FCM consumption. An original methodology was used, applicable for another future similar researches. The materials of the thesis can serve as methodological-didactic support in the university and postgraduate training of the medical staff, as well as for the preparation of the didactic materials (courses, guidelines) for the practitioners. The sources of Hg pollution were identified and characterized and their potential impact on the population's health was estimated, which created the possibility to argue, develop prophylactic measures specially to prevent exposure of children.

The applicative value of the research. The study allows the development, planning of measures and recommendations for the state public health surveillance service in order to recognize and address the mercury as a public health issue. The results of the study will serve as support for the practical departments of the NAPH in honouring the obligations and responsibilities stated in articles 16, 18, 19, 20, 21 of the Law no. 51 of 30.03.2017 on the ratification of the MC on mercury. The results of the study allowed the elaboration of the guidance material "*Guide on mercury incidents management: remedial options*" for Central and Local Public Authorities, Preschool and School Institutions, Public Medical Institutions.

Sharing and approval of research findings. The thesis was discussed, approved and recommended for defence at the joint meeting of the PhD supervisors, members of the guidance committee and the primary research unit (Scientific Laboratory of Chemical Dangers and Toxicology within NAPH) from 01.07.2021 (minutes no. 1), at the meeting of the Scientific Profile Seminar within Nicolae Testemitanu" State University of Medicine and Pharmacy of the Republic of Moldova on profile 331. Public Health, specialty Hygiene 331.02 of 17.09.2021 (minutes no. 1). Subsequently the thesis was recommended for public support by the decision of the Scientific Council of the Consortium from 02.12.2021 (No. 24). In order to carry out the study, the positive opinion of the Research Ethics Committee from 03.06.2016 no. 64 was granted.

Published articles. The research results were published in 16 scientific papers, 5 as single author, 1 article in an international scientific journal, 4 in accredited national scientific journals, 5 articles in the papers of scientific conferences, and 5 participations at scientific forums.

Keywords: mercury, methylmercury, estimated daily intake, fish, crustaceans and molluscs' consumption, mercury concentration, population's health risk, environment.

THESIS CONTENT

1. RESEARCH METHODOLOGY

1.1. Materials and methods of the research

The study was started in 2015 and finalized in 2020. It has been carried out in the scientific laboratory of Chemical Hazards and Toxicology of the NAPH. The Hg content in atmospheric air, soil, water, FCM was analysed and the risk assessment for the population's health was performed. It's a descriptive ecologic and analytic study focused on identification and characterization of Hg as a potential risk and the level of impact on human health by generating some hypotheses. The level of exposure was estimated for the general population and children aged 1-3 years, 3-7 years, and 7-18 years.

Currently, there is no a unified and standardized methodology for assessing the impact of Hg (metallic, MeHg) on the population's health [14, 17, 32, 34, 35]. Therefore, the use of available data from the literature as well as the own, original data was considered rational. The WHO methodological guide recommends at a first stage the application of the cost-effectiveness principle of research on assessing the level of exposure to Hg [40, 42].

To estimate the Hg concentration, samples of FCM (n = 220), soil (n = 200) and drinking water (n = 74) were collected, which were analysed in the Central Reference Sanitary-Hygienic Laboratory of NAPH by Atomic Absorption Spectroscopy method, with the technical support of the Food and Agriculture Organization of the United Nations (FAO).

The soil samples (200 samples) were collected by the grid collection method, dividing the territory of the RM into 74 equal squares, of 900 km² each. From the open areas (agriculture soils) 168 samples were collected, including: (i) North - 54 samples, (ii) Centre - 64 samples, (iii) South - 50 samples. In addition, 32 soil samples were collected from inside the cities (kindergartens). The samples were taken at a depth of 0-10 cm, with a stainless-steel spatula, in polypropylene bags, sealed, labelled and double-packed. Water samples (74 samples) were collected in polypropylene jars, sealed and labelled, including of artesian origin - 20 samples, from centralized sources - 35 samples and 19 water samples from springs, wells in areas adjacent to national roads.

Data on Hg concentration in the atmospheric air for the years 2014-2015 were taken from the European Emissions Monitoring Program (EMEP). The analysis of the Hg concentration in the atmospheric air was performed in comparison with Ukraine, Romania, EMEP [28].

Medical thermometers and lighting objects that contain Hg were classified according to the international classification system HS 2017 [23].

For Hg total content assessment in 26 species, 220 FCM coded samples for Dp were collected from the central Chisinau market and specialized stores, each with a mass of 100 g, packed in sterile polyethylene zip-lock packages. The transport time to the laboratory did not exceed six hours since the moment of collection. Auxiliary data from international databases (IDB) derived from GEMS/FOOD - Global Environmental Monitoring program was used [13, 41]. Relevant study data were extracted using the following filters: (i) *WHO Region(s)*: *all*; (ii) *Contaminant(s)*: *mercury, mercury (inorganic), methylmercury*; (iii) *Food Category(s)*: *Fish and other seafood (including amphibians, reptiles, snails and insects)*; (iv) *Food Name*: ALL; v) *Sampling Period*: 01.01.1972 -31.12.2018. We got 50331 recordings where 386 recordings were ruled out as incomplete data, others 1968 were excluded by the criteria that the FCM species were not consumed in the RM. For 21480 recordings out of 47977 left the measurement unit "µg/kg" was converted to "mg/kg". Recordings reported as being equal to the limit of detection (LOD) for

crustaceans - 15,1% (295), fish – 4,3% (1120), molluscs – 23,7% (1222) were adjusted using the formula recommended by [8, 9]:

 $X = LOD/\sqrt{2}$, where:

(1)

X – adjusted value,

LOD – Limit of Detection.

Hg concentrations in FCM species from the two data sources for the study (IDB, Dp) were compared by applying the Mann-Whitney statistical test, then merged into a single data set.

Consumption of FCM at national level for the period of 13 years (2005-2017) per species was extracted from FishStat J, (v.2017) [21], applying the following algorithm: (i) *Workspace (s)*: *FAO Global Fishery and Aquaculture Commodities Statistics and FAO Global Fishery and Aquaculture Production Statistics*; (ii) *Country: Republic of Moldova*; (iii) *Commodity(s)*: *Data extracted based on Harmonized Commodity Description and Coding Systems (HS-2017codes) & International Standard Statistical Classification of Aquatic Animals and Plants (ISSCAAP group)*; (iv) *Trade flow (s)*: *Imports, exports, re-exports, aquaculture and capture production*; (v) *Period: 2005-2017 (13 years)*.

Thence, 401 series were generated divided into 10 groups and 140 subgroups (HS-2017), were removed 59 series being reported as missing data for 2005-2017 and 14 series as: (i) *inedible*, (ii) *intended for animals*, (iii) *unfit for human consumption*. Volumes in tonnes, the quantity of FCM intended for consumption "*apparent consumption*" [21] was calculated by summing the quantities of FCM produced in the country plus the quantity imported with adjustments of stock changes and minus exports. In addition to real consumption, apparent consumption actually includes losses at storage, post-harvest, processing and packaging, distribution and does not represent the real quantity available for consumption by the final consumer. Liana et al. mentioned that apparent consumption overestimates the actual consumption of FCM by 44% [27]. Thereby, conversion factors were applied for each species based on the group. Conversion factor of 1 was assumed for groups 0304, 0305, 1504, 1604, 1605 as the quantities of FCM are presented as eviscerated, prepared, deboned, decapitated, etc. Consumption in grams per capita per day was deducted by dividing the total amount to the number of the population and 365 days a year, after the conversion factors were applied, assuming that 1 ton = 1000000 grams.

The yearbooks of the National Bureau of Statistics (NBS), Household Budget Surveys "*Aspects of the standards of living of the population*" for the years 2006-2018 were also used to assess the consumption of FCM by categories, such as: (i) by areas and regions (urban, rural, North, Centre, South, Chisinau); (ii) by the number of children in the household (one child, two children,> 3, no children); (iii) by household size (1 person, 2, 3, 4, 5 and more people per family); (iv) by socio-economic status (farmers, agricultural workers, non-agricultural workers, entrepreneurs, pensioners, others); (v) on quintiles (I, II, III, IV, V) [4].

The FCM consumption for children were considered Daily Required Quantity (DrQ) and Daily Consumed Quantity (DcQ) from "Recommendations for a healthy diet and tolerable physical activity in educational institutions from Republic of Moldova" (Order No. 638 of 12.08.2016) [29].

For data analysis, both those collected through experimental (direct) method and the bibliographic analysis (indirect), a series of descriptive (univariate, multivariate) and mathematical statistical methods were applied to identify the proportions, percentages, rates, frequency distributions or central trend measurement like, mean (\overline{x}), median (m_{dn}), modulus, deciles, quartiles, percentiles (p.), estimation of data scattering such as: standard deviation (SD),

coefficient of variation (CV). Whilst, different statistical tests, where appropriate, were applied to compare the obtained data, such as: unidirectional ANOVA test, Student's t test for equal and unequal variations, Leven's test for variation testing, Mann Whitney statistical test. These operations were performed using EPInfo 17, IBM SPSS statistics 26, Microsoft Excel 2016.

1.2. Special methodology for estimating risk and exposure to mercury

The Indirect Risk Assessment Method with a tiered approach is applied for the first time in the Republic of Moldova as a model for assessing the exposure risk to chemicals (Hg), which consists in exposure assessment by measuring the Daily Intake (DI) of Hg and MeHg for the general population and compare with safety limit values (RfD). The following formulas were used:

$$DI_{[HgS]} = \frac{2(C^{*}C)}{b.w.} *1000 [1];$$
(2)

(3)

(4)

(5)

- $DI_{[HgS]}$ =Daily intake by soil ingestion, (µg/kg b.w.-day)
- C = Hg concentration in the soil, (mg/kg)
- U = soil ingested amount for children 200 mg/day, adults 100 mg/day, [1, 13]
- b.w. = consumer average body weight (kg)
- 1000 = transformation coefficient from mg/kg to μ g/kg

$$DI_{[HgA]} = \frac{2(C+0)}{hw} * 1000 [1];$$

- $DI_{[HgA]} = Daily$ intake by water ingestion (µg/kg b.w.-day)
- C = Hg concentration in the water, (mg/l)
- U = the amount of water consumed for children 1 l/day, adults 2 l/day, [1, 13]
- b.w. = consumer average body weight (kg)
- 1000 = transformation coefficient from mg/kg to μ g/kg

 $DI_{[HgAr]} = \frac{\sum (C^*U)}{b.w.} *1000 [1];$

- $DI_{[HgAr]}$ = Daily intake by air, (µg/kg b.w.-day)
- C = Hg concentration in the air, (mg/m^3)
- U = inspired volume for children 12 m³/day, adults 15,2 m³/day, [1, 13]
- b.w. = consumer average body weight (kg)
- $1000 = \text{transformation coefficient from mg/kg to } \mu g/kg$

The $DI_{[Hg]}$ calculation which means the summary ingestion of Hg by soil, water, air was done using the formula:

$$\sum DI_{[Hg]} = DI_{[HgS]} + DI_{[HgA]} + DI_{[HgAr]}; (5)$$

 $DI_{[MeHg]} \text{ appraisement for FCM consumption was performed using the following formula:}$ $DI_{[MeHg]} = \frac{\sum (C_{(MeHg)}^{*U})}{\text{m.c.}}; [1, 17, 19, 42]$ (6)

- $DI_{[MeHg]}$ = Daily intake, for all consumed species, ($\mu g/kg b.w.-day$)
- $C_{(MeHg)} = MeHg$ concentration in analysed species, (mg/kg)
- U = Quantity of FCM consumed, (g/day);
- b.w. = consumer average body weight (kg)

For the evaluation of the Hg exposure, were considered the average body weight (b.w.) values recommended by EFSA: (i) the general population (per capita b.w.) - 60 kg; (ii) nursery (1-3year old) - 11,9 kg; (iii) kindergarten (3-7 years) - 22,3 kg; (iv) school (7-18 years) - 44,3 kg [17].

FCM consumed by children, the "U" value in formula (6) was recalculated using the DrQ values specified in the Order No. 638 dated by 12.08.2016, for boarding schools (7-18 years) - 110 g/day, kindergarten - age 3-7 years with an activity schedule of 9,5-10 hours - 45 g/day, nursery

children of 1-3 years with an activity schedule of 9,5-10 hours - 20 g/day [29]. DrQ values were divided into 10 equal thresholds, for example for 110 g/day the threshold of 11 g/day (110/10) was considered, then extracted the following thresholds: 11 g/day, 22 g/day, 33 g/day ... 110 g/day.

The exposure assessment level of MeHg among population categories was performed based on NBS data for the years 2006-2018 using the average values of FCM consumption and the values of the percentiles of MeHg concentration in FCM. The average consumption values were distributed by species, assuming that the consumption distribution per species for those categories is the same as the for the general population [6] estimated from FishStat J software, with the conversion from average kilograms per person per year to g/day and adjusted to FishStat J data, to be able to compare DI_[MeHg] with per capita values by applying a conversion factor of (0,496/ 49,6%) because NBS reports do not take into account food losses [4, 7, 27].

To calculate the characteristic risk of mercury, a non-carcinogenic substance, the Hazard Index (HI) was used, according to the formula:

$$HI = \frac{DI}{RfD} * 100\%; \tag{7}$$

- HI = Hazard Index (%);
- DI = Daily Intake ($\mu g/kg b.w.-day$);
- RfD = Reference Dose (μ g/kg b.w.-day)

The screening value (Hg^{Sv}_{PCM}), the maximum level of MeHg in the FCM, which allows a safe consumption of FCM (g/day) with no harm to health throughout all life, was calculated assuming that the RfD = 0,1 μ g/kg b.w. per day will not be exceeded:, formula (8) [12, 14]:

 $Hg_{PCM}^{Sv} = \frac{RfD * b.w.}{U_{r}}, \text{ where:}$ (8)

- Hg_{PCM}^{Sv} = Screening value (mg/kg)

- RfD = Reference Dose
$$(0.1 \mu g/kg b.w. - day)$$

- U_r = Recommended/consumed fish quantity (g/day) [29]
- b.w. = consumer average body weight (kg)

Another indicator is the Theoretical Allowed Consumption Rate (RtCa) (formula 9), which estimates the maximum amount of fish that can be consumed daily with a concentration (x) of MeHg, by a person with a certain b.w. so that during the life the RfD is not exceeded [12, 14]. $R_tC_a = \frac{RfD^*b.w.}{C}$, where: (9)

- R_tC_a = Theoretical Allowed Consumption Rate (g/day)
- b.w. = consumer average body weight (kg)
- C = MeHg concentration in the analysed FCM product (mg/kg).

The maximum number of FCM meals allowed to be consumed to reach RfD for MeHg was calculated and compared to the number of meals required to meet the recommended DHA+EPA polyunsaturated fatty acids intake. Estimates were made for four age groups: 1-3 years, 3-7 years, 7-18 years and per capita. The weight of one meal was assumed to be 52 g for 1-3 years, 70 g for 3-7 years, 101 g for 7-18 years and 120 g per capita [13].

The concentration of Hg in blood and hair was calculated based on the toxicokinetic model: $S_{HgT} = \frac{0.95*0.059*DI_{[MeHg]} *b.w.}{0.014 \ days^{-1}*Vs}; \text{ where: [40]}$ (10)

- S_{HgT} = Total Hg concentration in blood (µg/l)

- 0.95 = fraction of total Hg absorbed from DI_[MeHg]

- 0,059 = the fraction of total Hg absorbed by the blood tissue

- $DI_{[MeHg]}$ = Daily Intake (µg/kg b.w.-day)
- Vs = blood volume (liters)
- $0,014 \text{ days}^{-1} = \text{elimination constant from the blood}$
- b.w. = consumer average body weight (kg)

The presence of mercury in the hair indicates a chronic exposure. The concentration of mercury in the hair (P_{HgT}) can be determined using the estimates of its concentration in the blood by applying Legrand et al. formula (11) [25]:

(11)

$$P_{HgT} = \frac{S_{HgT}^{*250}}{1000}$$
, where: (8)

- P_{HgT} = Mercury concentration in hair (mg/kg)
- S_{HgT} = Total Hg concentration in blood (µg/l) based on formula (10)
- 250 = conversion factor (hair/blood)
- 1000 = adjustment factor for the measurement units, from μg to mg.

2. ESTIMATION OF METALLIC MERCURY EXPOSURE LEVEL

Metallic mercury is very stable and accumulates in the aquatic biota unless it is biotransformed by living organisms (methylation) [3, 15, 20, 38, 39, 42, 43].

The structure, distribution (%), emission quantities were studied in order to identify potential sources of mercury exposure. The National Inventory of Mercury Releases in the Republic of Moldova - 2014: Level 2 from 2017 was used for the emissions analysis [33]. We identified that in 2014 in the RM the total amount of releases is 972,12 kg Hg/year. The most important source of emissions is the group of consumer products with the intentionally use of mercury: 411,83 kg Hg/year (42,4% out of total), followed by waste storage/disposal and wastewater treatment: 270,89 kg Hg / year, (27,9%), then waste incineration: 101,32 kg Hg/year (10,4%), and other intentional uses of products/processes with 39,19 kg Hg/year (4,0%). 31,81% or 309,24 kg of total emissions are directed to general waste, 30,42% (295,8 kg) are emitted into the air, into soil 16,93% (164,58 kg) in terrestrial water basins 10,14% (98,67 kg) and 9,54% (92,82 kg) are recycled.

Therefore, we identified that during period of 2008-2018 on average 167391 thermometers per year with a decreasing tendency of 3641 units were imported, most of them being imported in 2010: 249787 units, the least in 2008: 83391 units (figure 1).



Figure 1. Import of Hg-based thermometers to the RM, 2008-2018 [23]

Thermometers were distributed to: medical facilities (11,60%), educational facilities and factories (2,58%), 85,82% among the population and other institutions [33, p.67-68] (figure 2).



Figure 2. Distribution per user of medical thermometers containing mercury

Lighting objects are considered another important source of Hg exposure among the population, especially when they are accidentally broken [33, p.74]. At least 3 types of lighting fixtures with Hg content were imported into the RM during 2001-2018 (figure 3). Total imported mercury or sodium vapor objects were 812377 units (code 853932), ultraviolet or infrared objects 1464046 units (code 853949) and 22111245 units (code 853931) discharge, fluorescent, hot cathode and double ended cap.



Figure 3. Import of lighting objects containing Hg, RM, 2001-2018 [23]

According to the NBS, the largest amount of toxic wastes - 3722 tons were formed in 2015, out of which 17 tons (0.45%) were transferred to household waste [5, 22]. In 2017, 1511 tons of toxic waste were produced, 621 tons (41%) being transferred to household waste. The average share of toxic waste transferred to household waste for 2001 - 2017 is $22,4\% \pm 22,6\%$ (SD).

The yearbooks of the State Ecological Inspectorate (IES) for 2014-2018 [22] in regional aspect (table 1), show that most waste landfills - 689 stretched over 530,98 ha are in the Centre region, occupying 0,049% of the area. In the North there are 641 waste landfills occupying 0,071% of the surface or 488,75 ha. At the country level, 1341,43 ha are covered with wastes (0,052%), distributed in 1860 waste landfills. Assuming that on average annually 22,4% \pm 22,6 (SD) of toxic wastes are transported to landfills then approximately 1730,39 \pm 1777,06 tons out of the total of 7865,39 tons of toxic waste are transferred to household wastes.

Region	Toxic	wastes (tones)		Landfills for household wastes				
	recorded	transferred to household wastes (x±SD)	population	n	area (ha)	% from total area		
North	756,84	166,505±171,046	955717	614	488,75	0,071		
Centre	5285,25	1162,76±1194,47	1831740,7	689	530,98	0,049		
South	1823,3	401,126±412,06	709994	557	321,7	0,037		
Country	7865,39	1730,39±1777,06	3497451,7	1860	1341,43	0,052		

Table	1. Some as	pects of waste	s storage, in	territorial	profile.	, 2014-2018	, RM	22
			A /			/	/ I	

EMEP concluded that current levels of Hg^0 in atmospheric air except for hot points are well below ML values [28]. The EEA (2017) and EU (2016) reports mentioned that the background levels of Hg in atmospheric air are 0,014 - 0,028 μ g/m³ and 0,07 μ g/m³ in urban areas [10, 15].

It was estimated that there is a significant difference between the average concentration of Hg⁰ (gas) in the air characteristic for the RM (\bar{x} =1,425, SD=0,089) compared to MSC-E data (\bar{x} =0,444, SD=0,686); t (69,816) = -84,382, p<0,001, or compared to Romania (\bar{x} =1,455, SD = 0,106); t (344) =2,09, p=0,037 (table 2). There is no significant difference between the average concentration of Hg0 (gas) characteristic for RM and Ukraine (\bar{x} =1,420, SD=0,094); t (734) =-0,355, p=0,723. All recorded values are below ML of 1,0 µg/m3 or 1000 ng/m³ [39].

Region	Forms	n	x [95%, CI]	SD	min/max	variation
	Hg(part)*	53244	40,595 [40,35:40,83]	28,61	0,007/90,0	89,993
MSC-E	Hg ⁰ (gas)	53244	0,444 [0,438:0,449]	0,686	0,0001/5,65	5,648
	Hg ²⁺ (gas)	53244	0,0118 [0,0117:0,0119]	0,0119	0,0002/0,806	0,806
Moldova	Hg ⁰ (gas)	62	1,425 [1,402:1,447]	0,089	1,3/1,587	0,287
Romania	Hg ⁰ (gas)	284	1,455 [1,443:1,467]	0,106	1,225/1,682	0,457
Ukraine	Hg ⁰ (gas)	674	1,420 [1,413:1,428]	0,094	1,225/1,880	0,656
		-	2			

Table 2. Concentrations of different forms of mercury in atmospheric air (ng/m³) [28]

Mark: * - the data are presented in picograms on m^3 ; ML – is considered 1000 ng/m³;

74 samples of drinking water were collected from different sources (table 3). The HgT concentration detected in all samples was below the LOQ of 0,0002 μ g/l and ML of 1,0 μ g/l. WHO in 2017 stated that Hg is present in inorganic form in surface and ground waters at concentrations usually below 0,5 μ g/l [43]. EEA, (2018) pointed that long-term pollution of water basins with Hg poses a risk of exposure to MeHg because metallic forms of Hg undergo the process of methylation further accumulating in FCM [1, 11, 16, 18, 19, 20, 38].

Table 3. Results of laboratory investigations of Hg levels in different water sources

Water sources	Collected samples	Concentration, (µg/l)	ML, (μg/l)		
Artesian	20				
Central	35		1.0		
Springs	19	<pre><loq (0,0002)<="" pre=""></loq></pre>	1,0		
Total	74				

Hg levels in agricultural soils (open areas) ranged from 0,001 mg/kg to 0,29 mg/kg (table 4). The mean values for each region were found to be 0,065 mg/kg [CI, 95%; 0,055:0,074] North, 0,072 mg/kg [CI, 95%; 0,053:0,091] at the Centre and 0,063 mg/kg [CI, 95%; 0,053:0,074] for the South. The mean value for RM is 0,049 mg/kg [CI, 95%; 0,041:0,057]. Applying the unidirectional ANOVA test (f (2,81) =0,477, p=0,622) it was found that there is no significant difference between regions. At the same time, no exceedances of ML (2,0 mg/kg) were detected.

Table 4. Characteristics of mercury concentration in soil samples in the RM

		n	x [95%, CI]	min/max	SD
Republic of Moldova	Centre	32	0,072 [0,053:0,091]	0,012/0,290	0,052
	Localities	32	0,001 [0,001:0,001]	0,001/0,001	0,000
	North	27	0,065 [0,055:0,074]	0,012/0,110	0,025
	South	25	0,063 [0,053:0,074]	0,019/0,103	0,025
Total		116	0,049 [0,041:0,057]	0,001/0,290	0,043

The most at risk are children with a potential cumulative mean exposure dose of 0,0149 μ g/kg b.w.-day compared to adults 0,0127 μ g/kg b.w.-day (table 5). The maximum cumulative values of DI_[Hg] to which children and adults could be exposed are equivalent to 0,0599 μ g/kg b.w.-day and 0,035 μ g/kg b.w.-day. Maximum HI is 1,5% for children and 0,88% for adults.

The WHO reports showed that exposure to Hg^0 through air, water and soil may be neglected in per capita exposure assessments, except for exposures at critical points with a high degree of pollution [37, 39, 42, 43].

Exposed	DI _[Hg] (µg/kg b.wday)								
group	soil (\overline{x}/max)	soil (\overline{x} /max)air (\overline{x} /max)watersummary (\overline{x} /max)							
Children	0,0134/0,058	0,0016/0,0012	0,00002	0,0149/0,0599	0,37/1,5				
Adults	0,0067/,029	0,0003/0,0003	0,0057	0,0127/0,035	0,31/0,88				

Table 5. Exposure of children and adults to metallic mercury

However, evidence based on own data and analysis of data from previous scientific researches provides evidence necessary for the initiation of policy actions on remediation and management of soil pollution (presence of numerous critical points) and mercury-containing devices (thermometers, lighting objects) [5, 15, 16, 20, 22, 24, 33, 38, 45].

3. ESTIMATION OF EXPOSURE TO MeHg THROUGH FCM CONSUMPTION

3.1. Characterization of MeHg concentration in FCM and FCM consumption

WHO stated that in order to calculate the estimated daily intake of MeHg it is necessary to have data on the amount of food consumed per person per day, the average concentration of MeHg in the analysed food and body weight of the estimated person for whom the exposure is estimated. Any change in one of these values leads directly to a change in the final exposure level [42].

To assess MeHg in FCM we combined two data sets with the total number (n = 33356

samples), including: (i) Dp (n = 220 samples) and (ii) IDB (n = 33136 samples) (figure 4).

The maximum average of MeHg concentration is in shark/swordfish – 0,823 mg/kg (SD: 0,75) and grenadier – 0,584 mg/kg (SD: 0,14). Hg concentrations in FCM are spread, as CV is> 0,3 and <1 for 8 species; > 1 for 18 species and only for 2 species, grenadier and sturgeon, CV is <0,3. We noted the presence of outliers (black lines) in 5 of 27 species, another 20 species contain both outliers and extreme values (red rings).



Figure 4. Box plot diagram regarding MeHg concentrations in different fish species

NBS reports that during the period of 2012-2016 in the country were imported FCM from 50 countries in the amount of 163695,1 thousand US dollars. Most were imported from: Norway 39851,8 thousand dollars (24,345%), Iceland – 29790,93 thousand dollars (18,2%), United Kingdom – 10826,69 thousand dollars (6,6%) and USA-10391,87 thousand dollars (6,35%) [7].

We estimated that the average per capita consumption for the years 2005-2017 was 19,68 g/day per capita, the most consumed species were: herrings – 6,463 g/day (32,8%), carp - 4,143 g/day (21,05%), unspecified species 3,091 g/day (15,7%), mackerel-1,609 g/day (8,17%), hake - 1,103 g/day (5,6%), whitening - 0,953 g/day (4,84%), salmon - 0,824 g/day (4,19%), cod - 0,745 g/day (3,78%), pollock with 0,224 g/day (1,14%), tuna - 0,09 g/day per capita (0,47%). All these listed species cover 97,3% of the average FCM consumption per capita (figure 5).



Figure 5. Share of individual species in total FCM consumption [21]

3.2. Exposure level to MeHg for different population groups

Therefore, taking into account the MeHg data concentrations dispersion and the distribution of FCM consumption (continuous growth trend) and the methodological aspects, we estimated the exposure for 10th, 20th, 30th, 40th, 50th, 60th, 70th, 80th, 95th percentiles (table 6).

וח		Percentiles assigned to FCM consumption										
	1 []~]	10	20	30	40	50	60	x	70	80	90	95
Inter	ngj	(14,2)	(15,3)	(16,1)	(17,3)	(18,2)	(18,9)	(19,68)	(20,8)	(24,2)	(27,4)	(29,2)
	10	0,003	0,004	0,004	0,004	0,005	0,005	0,005	0,006	0,007	0,008	0,009
g	20	0,005	0,006	0,006	0,007	0,007	0,008	0,008	0,008	0,010	0,012	0,013
de F	30	0,007	0,008	0,008	0,009	0,009	0,010	0,010	0,011	0,013	0,016	0,017
ned to N ation	40	0,009	0,010	0,010	0,011	0,012	0,013	0,013	0,014	0,017	0,020	0,022
	50	0,012	0,013	0,014	0,015	0,016	0,017	0,018	0,019	0,022	0,027	0,029
sig	60	0,016	0,018	0,019	0,020	0,022	0,023	0,024	0,026	0,030	0,036	0,039
s as	70	0,022	0,024	0,026	0,028	0,030	0,032	0,033	0,035	0,042	0,050	0,053
tile cc	x	0,022	0,025	0,027	0,029	0,031	0,032	0,034	0,036	0,043	0,051	0,054
Percen	80	0,031	0,035	0,037	0,040	0,043	0,045	0,047	0,050	0,060	0,071	0,076
	90	0,051	0,056	0,061	0,066	0,069	0,073	0,076	0,081	0,097	0,114	0,121
	95	0,075	0,084	0,091	0,098	0,103	0,109	0,114	0,120	0,143	0,169	0,179

Table 6. Distribution of dietary exposure (per capita) to MeHg and the Hazard Index (HI)

Notes: green cells represent $DI_{[MeHg]}$ values between 0 and 0,0899 µg/kg b.w.-day, 0<HI <0,899, safe area; yellow cells represent $DI_{[MeHg]}$ values between 0,09-0,0999 µg/kg b.w.-day, 0,899<HI <0,999, risk area; red cells: $DI_{[MeHg]} => 0,1 \mu g/kg$ b.w.-day, HI>1,0.

We estimated that $DI_{[MeHg]}$ per capita varies from 0,003 µg/kg b.w.-day (HI – 0,03) to 0,179 µg/kg b.w.-day (HI - 1,79). The $DI_{[MeHg]}$ average is 0,034 µg/kg b.w.-day for a consumption rate of 19,68 g/day of FCM. The $DI_{[MeHg]}$ median is 0,016 µg/kg b.w.-day (HI - 0,16). In 9 cases out of 121 estimated values the $DI_{[MeHg]}$ is above the reference value (red cells; HI ≥ 1).

Table 7. Dieta	ry exposure to	• MeHg and HI	for boarding school	l children (7-18 vears)
	v 1		8	

			Concentration percentiles										
tion (g/day)		10	20	30	40	50	60	70	x	80	90	95	
	11	0,004	0,006	0,008	0,010	0,014	0,019	0,026	0,026	0,037	0,059	0,088	
	22	0,008	0,012	0,016	0,021	0,027	0,037	0,052	0,053	0,073	0,118	0,176	
	33	0,012	0,018	0,024	0,031	0,041	0,056	0,077	0,079	0,110	0,177	0,264	
	44	0,016	0,025	0,032	0,042	0,055	0,074	0,103	0,105	0,147	0,236	0,352	
	55	0,020	0,031	0,041	0,052	0,069	0,093	0,129	0,131	0,183	0,295	0,440	
dur	66	0,025	0,037	0,049	0,063	0,082	0,112	0,155	0,158	0,220	0,354	0,528	
ISUC	77	0,029	0,043	0,057	0,073	0,096	0,130	0,180	0,184	0,256	0,413	0,616	
Ŭ	88	0,033	0,049	0,065	0,084	0,110	0,149	0,206	0,210	0,293	0,472	0,704	
	99	0,037	0,055	0,073	0,094	0,124	0,167	0,232	0,237	0,330	0,531	0,792	
	110	0,041	0,061	0,081	0,104	0,137	0,186	0,258	0,263	0,366	0,591	0,879	

Notes: green cells represent $DI_{[MeHg]}$ values between 0 and 0,0899 µg/kg b.w.-day, 0<HI <0,899, safe area; yellow cells represent $DI_{[MeHg]}$ values between 0,09-0,0999 µg/kg b.w.-day, 0.899<HI <0,999, risk area; red cells: $DI_{[MeHg]} => 0,1 \mu g/kg b.w.-day$, HI>1.0.

For children aged 7-18 (table 7) who are enrolled in boarding schools $DI_{[MeHg]}$ varies from 0,004 µg/kg b.w.-day (consumption = 11 g/day; MeHg concentrations equivalent to percentile 10) to 0,879 µg/kg b.w.-day (HI - 8,79) (consumption =110 g/day; MeHg concentrations equivalent to percentile 95). For average MeHg concentrations, the $DI_{[MeHg]}$ is 0,026 µg/kg b.w.-day (HI - 0,26) for a consumption of 11 g/day and 0,263 µg/kg b.w.-day (HI - 2,6) for a consumption of 110 g/day.

For kindergarten children with 9,5 - 10 hours schedule, the $DI_{[MeHg]}$ varies from 0,003 µg/kg b.w.-day to 0,676 µg/kg b.w.-day (HI - 6,76) (table 8). Considering DrQ of 45 g/day and average MeHg concentrations then estimated $DI_{[MeHg]}$ is 0,202 µg/kg b.w.-day (HI - 2,02). At a consumption shortage of 40% (DcQ) or 27 g/day, the $DI_{[MeHg]}$ is 0,121 µg/kg b.w.-day (HI - 1,21).

			20 30 40 50 60 70 x 80 90 95 3 0,005 0,006 0,008 0,011 0,014 0,020 0,020 0,028 0,045 0,068 6 0,009 0,012 0,016 0,021 0,029 0,040 0,040 0,056 0,091 0,135 9 0,014 0,019 0,024 0,032 0,043 0,059 0,061 0,084 0,136 0,203 3 0,019 0,025 0,032 0,042 0,057 0,079 0,081 0,113 0,182 0,270 6 0,024 0,031 0,053 0,071 0,099 0,101 0,141 0,227 0,338										
		10	20	30	40	50	60	70	X	80	90	95	
	4.5	0,003	0,005	0,006	0,008	0,011	0,014	0,020	0,020	0,028	0,045	0,068	
()	9	0,006	0,009	0,012	0,016	0,021	0,029	0,040	0,040	0,056	0,091	0,135	
/da	13.5	0,009	0,014	0,019	0,024	0,032	0,043	0,059	0,061	0,084	0,136	0,203	
ı (g	18	0,013	0,019	0,025	0,032	0,042	0,057	0,079	0,081	0,113	0,182	0,270	
otio	22.5	0,016	0,024	0,031	0,040	0,053	0,071	0,099	0,101	0,141	0,227	0,338	
dun	27	0,019	0,028	0,037	0,048	0,063	0,086	0,119	0,121	0,169	0,272	0,406	
Const	31.5	0,022	0,033	0,044	0,056	0,074	0,100	0,139	0,141	0,197	0,318	0,473	
	36	0,025	0,038	0,050	0,064	0,085	0,114	0,158	0,162	0,225	0,363	0,541	
	40.5	0,028	0,042	0,056	0,072	0,095	0,129	0,178	0,182	0,253	0,409	0,608	
	45	0,031	0,047	0,062	0,080	0,106	0,143	0,198	0,202	0,282	0,454	0,676	

Table 8. Dietary exposure to MeHg and HI for children in kindergarten (3-7 years)

Notes: green cells represent $DI_{[MeHg]}$ values between 0 and 0,0899 μ g/kg b.w.-day, 0<HI <0,899, safe area; yellow cells represent $DI_{[MeHg]}$ values between 0,09-0,0999 μ g/kg b.w.-day, 0,899<HI <0,999, risk area; red cells: $DI_{[MeHg]} => 0,1 \ \mu$ g/kg b.w.-day, HI>1,0.

Children of 1-3 years are exposed to MeHg between 0,003 μ g/kg b.w.-day (HI-0,03) at 2 g/day of FCM and 0,583 μ g/kg b.w.-day (HI - 5,83) for 20 g/day (table 9).

			Concentration percentiles10203040506070\$\overline{x}\$8090950,0030,0040,0050,0070,0090,0120,0170,0170,0240,0390,0580,0050,0080,0110,0140,0180,0250,0340,0350,0490,0780,1170,0080,0120,0160,0210,0270,0370,0510,0520,0730,1170,1750,0110,0160,0220,0280,0360,0490,0680,0700,0970,1570,2330,0140,0200,0270,0350,0460,0620,0850,0870,1210,1960,2920,0160,0240,0320,0420,0550,0740,1030,1050,1460,2350,3500,0190,0280,0380,0480,0640,0860,1200,1220,1700,2740,4080,0220,0330,0430,0550,0730,0990,1370,1390,1940,3130,4670,0240,0370,0480,0620,0820,1110,1540,1570,2190,3520,525									
on (g/day)		10	20	30	40	50	60	70	$\overline{\mathbf{X}}$	80	90	95
	2	0,003	0,004	0,005	0,007	0,009	0,012	0,017	0,017	0,024	0,039	0,058
y)	4	0,005	0,008	0,011	0,014	0,018	0,025	0,034	0,035	0,049	0,078	0,117
ı (g/day	6	0,008	0,012	0,016	0,021	0,027	0,037	0,051	0,052	0,073	0,117	0,175
	8	0,011	0,016	0,022	0,028	0,036	0,049	0,068	0,070	0,097	0,157	0,233
otio	10	0,014	0,020	0,027	0,035	0,046	0,062	0,085	0,087	0,121	0,196	0,292
ımpı	12	0,016	0,024	0,032	0,042	0,055	0,074	0,103	0,105	0,146	0,235	0,350
Isuc	14	0,019	0,028	0,038	0,048	0,064	0,086	0,120	0,122	0,170	0,274	0,408
Cc	16	0,022	0,033	0,043	0,055	0,073	0,099	0,137	0,139	0,194	0,313	0,467
	18	0,024	0,037	0,048	0,062	0,082	0,111	0,154	0,157	0,219	0,352	0,525
Consumption (g/day)	20	0.027	0.041	0.054	0.069	0.091	0.123	0.171	0.174	0.243	0.392	0.583

Table 9. Dietary exposure to MeHg and HI for nursery children (1-3 years)

Notes: green cells represent $DI_{[MeHg]}$ values between 0 and 0,0899 $\mu g/kg$ b.w.-day, 0<HI <0,899, safe area; yellow cells represent $DI_{[MeHg]}$ values between 0,09-0,0999 $\mu g/kg$ b.w.-day, 0,899<HI <0,999, risk area; red cells: $DI_{[MeHg]} => 0,1 \ \mu g/kg$ b.w.-day, HI>1,0.

3.3. Recommendations for the consumption of fish, crustaceans and molluscs

Species like bivalve molluscs, herrings, salmon, trout, cephalopods, crustaceans, pollack, anchovies and haddock are the most optimal option because they can be consumed more than 2 servings per week (s/w) regardless of age (table 10).

		-								
Species	Maxin	num allow week to r	able servi each RfD	ngs per	Serves per week to fulfil EPA + DHA recommendations					
species	1-3	3-7	7-18	per	1-3	3-7	7-18	per		
	years	years	years	capita	years	years	years	capita		
Bivalve molluscs	5,2	7,5	9,8	11,4	5,9	8,8	6,1	5,1		
Herring	4,3	6,1	8,0	9,3	0,8	1,3	0,9	0,7		
Salmon	4,0	5,7	7,4	8,6	0,8	1,2	0,8	0,7		
Trout	2,8	4,1	5,3	6,2	0,9	1,3	0,9	0,8		
Cephalopods	2,8	4,0	5,2	6,1	3,8	5,7	3,9	3,3		
Crustaceans	2,5	3,7	4,8	5,5	4,1	6,1	4,2	3,5		
Pollock	2,3	3,4	4,4	5,1	6,7	10,0	6,9	5,8		
Anchovies	2,2	3,2	4,2	4,9	1,2	1,7	1,2	1,0		
Haddock	2,2	3,2	4,2	4,9	7,1	10,5	7,3	6,1		
Cod	1,9	2,8	3,7	4,3	8,0	11,9	8,3	6,9		
Sturgeon	1,8	2,6	3,4	4,0	2,0	3,0	2,1	1,7		
Bream sea	1,6	2,3	3,0	3,5	1,9	2,8	2,0	1,7		
Carp	1,5	2,2	2,9	3,3	3,7	5,5	3,8	3,2		
Redfish	1,5	2,2	2,8	3,3	2,0	3,0	2,1	1,8		
Mackerel	1,2	1,7	2,3	2,6	0,5	0,8	0,5	0,5		
Whitening	1,2	1,7	2,3	2,6	14,0	20,8	14,4	12,2		
Eel	0,9	1,3	1,7	1,9	0,9	1,3	0,9	0,8		
Pike/perch	0,9	1,2	1,6	1,9	6,3	9,3	6,5	5,4		
Unspecified	0,8	1,2	1,6	1,8	2,1	3,1	2,2	1,8		
Flat fishes	0,8	1,2	1,5	1,8	4,6	6,8	4,7	4,0		
Hake	0,8	1,1	1,5	1,7	11,8	16,6	11,5	9,7		
Ocean perch	0,8	1,1	1,5	1,7	1,7	2,6	1,8	1,5		
Tuna	0,5	0,5	1,0	1,2	2,2	3,2	2,3	1,9		
Ling	0,5	0,7	0,9	1,1	10,8	15,2	10,5	8,8		
Grenadier	0,3	0,4	0,5	0,6	14,0	20,8	14,4	12,2		
Sharks	0,2	0,3	0,4	0,4	2,5	3,7	2,5	2,1		
General	1,5	2,2	2,9	3,4						

Table 10. Number of weekly servings needed to rich RfD for MeHg comparatively with number of needed servings to fulfil the standards recommended for EPA + DHA

Note: green cells - species allowed to be consumed >2 s/w (best choice); yellow cells: 0,8 - 1,9 s/w; red cells < 0,5 - try to avoid them.

Other two species: shark/swordfish and grenadier are recommended to be avoided, maximum <0,5 s/w regardless of age [12]. Of the 26 species analysed, only herrings could be consumed a maximum of 4.3 s/w (31,6 g/day) by children in the nursery, 6,1 s/w (61,3 g/day) by children in kindergartens, 8,0 s/w (115,2 g/day) of boarding school children, per capita could be consumed 9,3 s/w (159,3 g/day). Salmon could be consumed a maximum of 4,0 s/w (29,3 g/day) by children in the nursery, 5,7 s/w (56,9 g/day) by children in kindergartens, 7,4 s/w (106,9 g/day)

by children from boarding schools, per capita 8,6 s/w (147,7 g/day).

Only 6 out of 26 species, assuming that the only source of DHA + EPA are FCM, could provide a sufficient daily intake of EPA + DHA polyunsaturated fatty acids for all age groups in respect to the maximum number of servings allowed per week, in terms of mercury content: herrings, salmon, trout, anchovies, mackerel and eel. Thus, to fulfil the 0,125 g/day of EPA + DHA for children in the nursery are needed 0,8 s/w (6,3 g/day) of herrings, 1,3 s/w (12,5 g/day) for children in kindergarten, for those in boarding schools – 0,9 s/w (12,5 g/ day), per capita – 0,7 s/w (12,5 g/day). Respecting the assortment and share of fish species consumed nationally, then for children in nurseries would be allowed to consume only 1,5 s/w or 11,4 g/day of fish, from kindergartens only 2,2 s/w or 22,3 g/day, from boarding schools – 2,9 s/w or 41,9 g/day, the general population 3,4 s/w or 57,9 g/day, without being exposed to MeHg over RfD. EFSA (2014) recommends a consumption of 1-2 servings of fish (equivalent to 150-300 g) per week for children aged 7-18 years and adults would be sufficient to ensure a necessary intake of Omega-3 essential fatty acids [18, 30].

4. TOXICOKINETICS MODEL AS A BIOMONITORING TOOL

Modelled concentrations of MeHg in blood ranged from 0,2 μ g/l to 11,04 μ g/l (table 11). The average value according to the estimated data of the concentration of MeHg in the blood is 2,09 μ g/l. The median concentration of MeHg in the blood is 0,99 μ g/l.

DI [MeH	[g]	Consumption percentiles											
		10	20	30	40	50	60	X	70	80	90	95	
	10	0,2	0,23	0,25	0,27	0,29	0,31	0,33	0,35	0,41	0,5	0,54	
	10	/0,05	/0,06	/0,06	/0,07	/0,07	/0,08	/0,08	/0,09	/0,1	/0,12	/0,13	
	20	0,32	0,35	0,37	0,4	0,44	0,47	0,49	0,52	0,61	0,74	0,8	
	$ \begin{array}{c} 1g] \\ 10 \\ 20 \\ 30 \\ 40 \\ 50 \\ 60 \\ 70 \\ \overline{x} \\ 80 \\ 90 \\ \end{array} $	/0,08	/0,09	/0,09	/0,1	/0,11	/0,12	/0,12	/0,13	/0,15	/0,18	/0,2	
	30	0,42	0,47	0,5	0,54	0,58	0,62	0,65	0,69	0,81	0,97	1,05	
	30	/0,11	/0,12	/0,12	/0,13	/0,15	/0,15	/0,16	/0,17	/0,2	/0,24	/0,26	
s	10	0,55	0,6	0,64	0,69	0,75	0,79	0,83	0,88	1,05	1,25	1,35	
tile	40 50	/0,14	/0,15	/0,16	/0,17	/0,19	/0,2	/0,2	/0,22	/0,26	/0,31	/0,34	
ercent	50	0,72	0,8	0,85	0,92	0,99	1,04	1,09	1,16	1,38	1,65	1,77	
		/0,18	/0,2	/0,2	/0,23	/0,25	/0,26	/0,27	/0,29	/0,35	/0,41	/0,44	
d u	60	0,97	1,08	1,15	1,25	1,34	1,4	1,48	1,57	1,87	2,22	2,39	
itio		/0,24	/0,27	/0,3	/0,31	/0,33	/0,35	/0,37	/0,39	/0,47	/0,56	/0,6	
ıtra	70	1,36	1,51	1,61	1,74	1,85	1,95	2,05	2,18	2,59	3,07	3,29	
cer	70	/0,34	/0,38	/0,4	/0,43	/0,46	/0,49	/0,51	/0,54	/0,65	/0,77	/0,82	
son	v	1,39	1,54	1,65	1,79	1,9	2	2,09	2,22	2,65	3,12	3,33	
0	А	/0,35	/0,39	/0,41	/0,45	/0,47	/0,5	/0,52	/0,56	/0,66	/0,78	/0,83	
	80	1,92	2,14	2,3	2,48	2,64	2,77	2,92	3,09	3,69	4,38	4,67	
	80	/0,48	/0,54	/0,57	/0,62	/0,66	/0,69	/0,73	/0,77	/0,92	/1,09	/1,17	
	00	3,12	3,47	3,74	4,04	4,26	4,48	4,7	4,98	5,94*	7,02*	7,47*	
	90	/0,78	/0,87	/0,93	/1,01†	/1,07†	/1,12†	/1,18†	/1,24†	/1,49†	/1,75†	/1,87†	
	95	4,64	5,19	5,6	6,05*	6,37*	6,69*	7,0*	7,42*	8,83*	10,4*	11,0*	
	95	/1,16†	/1,3†	/1,4†	/1,51†	/1,59†	/1,67†	/1,75†	/1,85†	/2,21†	/2,6†	/2,76†	

Table 11. Modelled concentration of MeHg in blood (µg/l) and hair (mg/kg)

Notes: * *Blood MeHg concentration* > 5,8µg/L; † *Hair MeHg concentration* >1,0 mg/kg;

We note that more studies (1, 14, 26, 25, 30, 40, 42) mentioned that the level of MeHg in the blood corresponding to a level of exposure equivalent to RfD (0,1 μ g/kg m.c. – day) is 5,8 μ g/l

and 1,0 mg/kg for the hair MeHg concentration. In other words, the concentration of MeHg in the blood would exceed the critical (safety) values for a person consuming FCM amounts greater than or equal to 24,2 g/day (80th percentile) provided that the concentration of MeHg in the species consumed is at least equal to or greater than the 80th percentile or if the FCM consumption is at least 17,3 g/day or more, but the MeHg concentration will be no lower than the 95th percentile value. Mercury in the hair among the general population is 0,52 mg/kg, median = 0,25 mg/kg.

CONCLUSIONS

- Concentrations of Hg⁰ (gas) in atmospheric air is 1,425 ng/m³ (95% CI, 1,402: 1,447), total Hg in soil 0,049 mg/kg, in drinking water total Hg was less than LOQ of 0,0002 μg/l. For water, air and soil there were no exceedances of the RfC value. The cumulative DI_[Hg] for children is 0,0599 μg/kg b.w.- day (HI – 1,5%), adults – 0,035 μg/kg b.w.- day (HI – 0,88%). Thus, exposure to the Hg from the environment can be characterized as "negligible risk".
- 2. In the period 2008 2018, 1841297 thermometer units were imported, 85,82% being purchased by the population, 11,60% by medical institutions and 2,58% distributed in educational institutions and enterprises. In the period 2001-2018 were imported 812377 units of mercury or sodium vapor lamps, 1464046 units of ultraviolet or infrared bodies, 22111245 units discharge, fluorescent, hot cathode, double head. The presence of these devices on the territory of the Republic of Moldova requires the strengthening of institutional capacities for their elimination which would reduce the premises for accidental exposures to Hg.
- 3. The presence of 1860 landfills are a source of exposure to Hg when $1730,39 \pm 1777,06$ tonnes (SD) per year of toxic wastes are transferred to household wastes. Contamination of these areas increases the risk of direct and indirect exposure to Hg of the population as most landfills are not guarded and only 186 (10%) landfills are authorised.
- 4. The average consumption of PCM for the period 2005-2017 was 19,68 g/day per capita, of which 97,3% being formed by clupeids (herring, sardines) 32,8%, carp 21,05%, unspecified species 15,7%, mackerel 8,17%, hake 5,6%, putasu 4,84%, salmon 4,19%, mackerel 3,78%, Polish 1,14%, tuna 0,47%.
- 5. It was estimated that DI_[MeHg] per capita varies from 0,003 μg/kg b.w.- day (HI = 0,03) to 0,179 μg/kg b.w.- day (HI = 1,79), the average 0,034 μg/kg b.w.- day (HI 0,3). DI_[MeHg] for boarding school children range from 0,004 μg/kg b.w.- day (HI 0,04) for a consumption of 11,0 g/day PCM to 0,879 μg/kg b.w.- day (HI 8,79) for 110 g/day. Those in kindergartens could be exposed from 0,003 μg/kg b.w.- day (HI 0,03) consuming 4,5 g/day of FCM to 0,666 μg/kg b.w.- day (HI = 6,76) if they consume 45 g/day of FCM. Children attending the nursery would be exposed between 0,003 μg/kg b.w.- day (HI 0,03) if they consumed 2 g/day of PCM and 0,583 μg/kg b.w.- day (HI 5,83) if they consumed 20 g/day.
- 6. For children only clupeids and salmon meet the safety criteria, in relation to the recommendations of the Order No.638 from 2016. Clupeids could be consumed a maximum of 31,6 g/day by those aged 1-3 years, 61,3 g/day by children aged 3-7 years, and 115,2 g/day by children aged 7-18 years. Salmon could be consumed a maximum of 29,3 g/day by children aged 1 3 years, 56,9 g/day by children aged 3 7 years, 106,9 g/day by children age 7-18 years.
- 7. Contamination of environmental factors with mercury and its presence in fish, crustaceans and molluscs remains a current public health problem in the Republic of Moldova due to the lack in the Republic of Moldova of an adequate, coordinated, integrated and dedicated

mercury monitoring system for air, water, soil, fish, crustaceans and molluscs. Thus, the assessment of the potential and actual risk of the influence of mercury on the health of the population must be included in the list of priority tasks of the public health surveillance service.

PRACTICAL RECOMMENDATIONS

- I. At the legislative and normative level:
 - 1. To strengthen the national regulatory framework of Hg as a useful substance and waste.
 - 2. Recalculation of consumption recommendations for FCM depending on the presence of mercury and other toxic substances, provided in the national legislation for children of different ages enrolled in preschools, schools.
- II. At the level of the Ministry of Health, Labour and Social Protection:
 - 1. Emphasis should be on adequate communication of the risks of exposure to Hg and the nutritional and socio-economic benefits of fish consumption, in particular in children food.
 - 2. Initiate an effective mercury monitoring program at national level to sample and determine key indicators of mercury concentration in air, water, soil, fish, crustaceans and molluses.
 - 3. For a complete, real and clear picture of the distribution of the population's exposure to MeHg and Hg at national and regional level, it is urgently necessary to assess the exposure, especially of children and the adult population with a high consumption of fish.
- III. At the level of IP USMF "Nicolae Testemitanu", ANSP, other specialized scientific institutions:
 - 1. Increasing the number of studies on mercury exposure, especially for children.
 - 2. Initiation of biomonitoring and screening studies to monitor methylmercury exposure trends especially for vulnerable populations (children).
 - 3. Subsequent studies on population exposure to MeHg should include an in-depth assessment of the consumption of FCM while collecting samples to determine the concentration of MeHg and estimating the possible exposure of pregnant women, young children and adults. Validation of the relationship between Hg concentration in blood and hair with FCM consumption.
 - 4. Characterize the intake of Hg in certain subpopulations by detailed assessment, based on specific information on local food habits and Hg levels, found in FCM consumed by local populations.
 - 5. Good practices exchange and updating training and re-training courses in the field of public health for students, residents and specialties based on national and international partnerships.
- IV. At the level of medical institutions:
 - 1. Strengthening the capacity of public health institutions in the diagnosis and treatment of mercury poisoning and its compounds.
 - 2. Training of family doctors, nutritionists and nurses in the field of prevention of mercury poisoning and its compounds.
- V. At the level of the individual:
 - 1. Promoting a healthy lifestyle through a healthy and balanced diet.
 - 2. Information from reliable sources on environmental pollution and food safety, reading nutrition labels; consulting public health specialists on the possible mercury contamination

of products, the rational level of consumption of food products likely to accumulate mercury; use of information platforms, etc.).

3. Carrying out a personal record of food consumption, allowing a daily self-monitoring by adults, pregnant women, and young children of the consumption of FCM to estimate the personal intake of mercury and take the necessary corrective measures.

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LIST OF THESIS PUBLICATIONS, PATENTS AND SCIENTIFIC FORUMS AT WHICH RESEARCH RESULTS WERE PRESENTED

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Articles in accredited national scientific journals: ✓ articles in category A journals

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