

Evaluation of Domestic Sources of Mercury



August 2000

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Association of Metropolitan Sewerage Agencies
August 2000

1.0 INTRODUCTION

The U.S. Environmental Protection Agency's (EPA's) actions to control and eliminate mercury sources to the environment emphasize the need for controlling point sources to publicly owned treatment works (POTWs). There have been claims made that elimination/minimization of mercury from industrial sources such as dental facilities and hospitals would result in "zero discharge" or at least enable POTWs to meet extremely low aquatic life and wildlife water quality criteria for mercury. In fact, in its proposed mixing zone ban for mercury and other bioaccumulative pollutants in the Great Lakes, EPA noted that there were several documented instances in the Great Lakes Basin and elsewhere where the development and implementation of aggressive source control programs had resulted in the virtual elimination of bioaccumulative pollutants.¹ This approach, however, assumes that there is no background mercury concentration in domestic wastes. Until recently there has been very little information on domestic waste concentrations, mainly due to a lack of monitoring at a sufficiently low level.

To better understand the relative contributions of mercury in domestic wastes and potential source control options, a study was initiated to collect information on concentrations of mercury in domestic wastewater, to identify the sources of mercury in domestic wastewater, and to evaluate the feasibility of controlling those mercury sources. This report presents the results of the investigation and recommendations on future courses of action.

2.0 DOMESTIC WASTEWATER DATA

When we began this project, we identified several AMSA members that had collected mercury data for residential wastewater using sensitive sampling and analytical methods. Our goal was to see what kinds of mercury concentration ranges existed in domestic wastewater without inputs from industry, dentists, or other commercial sources. The participating agencies were geographically distributed (east coast, midwest, west coast) and represented different sizes and types of POTWs. It should be noted that while the purpose of this evaluation was not to provide a statistically valid national sample, it was considered to be statistically *significant* as a national sample set to provide insight on solutions for regulating mercury in wastewater discharges.

Domestic wastewater data presented in this report were collected by the following four POTWs: the Massachusetts Water Resources Authority (MWRA) in Boston, Massachusetts, the Metropolitan Council Environmental Services (MCES) in St. Paul, Minnesota, the Hampton Roads Sanitation District (HRSD) in Virginia Beach, Virginia and the City of Palo Alto, California. The sampling locations were carefully screened in all areas to ensure that only residential wastewater flows were sampled with no commercial or industrial inputs. Age of the sampled residential service areas ranged from <10 to ~125 years old.

¹ October 4, 1999 *Federal Register*, pp. 53641-53642.

Mercury data were produced using analytical methods sensitive enough to generate results that could be compared to the current EPA water quality criteria. Statistical evaluations were performed on the quantifiable data. Some of the samples were collected and analyzed using clean techniques. The remaining data were collected using semi-clean protocols with measures employed to control contamination.

2.1 Scope of Data Evaluation

- Three hundred and eighty three (383) data points were obtained from samples collected from residential areas; of these, 263 had detectable results based on the detection limits (MDLs) of the analytical methods used by the participating POTWs.
- The list of analytical methods used and the corresponding MDLs are presented in Table 1.
- Mean and median mercury concentrations for each residential neighborhood/service area and for all areas combined were calculated using all detectable results.
- Since a few of the results appeared to be outliers, a test was performed to determine which data points were indeed statistical outliers. The calculations of mean and median values were then performed excluding from the data sets those few measurements that were determined to be outliers.
- Additional statistical calculations were performed with inclusion of all detectable and non-detectable values, with non-detectable values reported as concentrations at the MDL.
- Trends in mercury concentration as related to an age of each residential area were assessed statistically.
- Seasonal changes, (on a quarterly basis) in mercury concentrations were also statistically evaluated.

Table 1. Summary of Analytical Methods and Detection Limits

POTW	Analytical Method	MDL – ng/L
MWRA	EPA 245.1	50
MCES	EPA 245.7	15
HRSD	EPA 245.7	2
Palo Alto	Hydride by FIAS/AA ¹	100
Palo Alto	Hydride by FIAS/Gold Amalgamation/AA ²	10
Palo Alto	Hydride by FIAS/ICP-MS ³	10
¹ Samples collected between 7/83-2/96		
² Samples collected between 3/96-8/97		
³ Samples collected between 9/97-Present		

2.2 Statistical Methods

- The data were normalized (\log_{10}) before statistical analyses were performed. Outliers were determined using Box-and-Whisker and Normal Probability Plots (*Statgraphics Plus for Windows 4.0* used for statistical evaluations).

- After normalizing the data, standard skewness and kurtosis values were obtained. (Skewness measures the symmetry or shape of the data. The flatness or steepness of the data distribution, with respect to a normal or Gaussian distribution, is measured by kurtosis). The results indicated that the data significantly departed from normality for the sets including outlier concentrations. Therefore, any statistical evaluations regarding standard deviation (e.g., ANOVA) tend to be invalidated.
- Normalized data sets excluding outliers returned standard skewness and kurtosis values indicating that the data had a normal distribution.
- Mean and median mercury values and standard deviations were calculated for each neighborhood and for all areas studied.
- Seasonal changes in mercury concentration were assessed using ANOVA on the normalized data set.
- The MCES data were generated on split samples by two different laboratories, using different analytical methods. The Student's t-test performed on these data sets indicated the sets were not statistically different, therefore a mean value for each sample split was used in the statistical evaluations. One sample split with the RPD of >100% was rejected.

2.3 Results

Table 2 presents a summary of all results, and also provides information on the approximate age of each neighborhood and average total suspended solids (TSS) concentrations, where available.

- Using all of the mercury data, the mean and median values for all of residential areas were:

Mean	178 ng/L
Median	110 ng/L

- The normalized data set, with an exclusion of extremely high values, which were considered outliers, provided the following mercury mean and median values:

Mean	138 ng/L
Median	104 ng/L

- Using the data sets that included non-detected values at the MDL concentrations, the mean and median mercury values for all of the residential areas were:

Mean	143 ng/L
Median	88 ng/L

Table 2. Summary of Mercury Concentrations for All Service Areas

Service Area	Number of Samples	Dates of Sampling	Age of area (yrs)	Avg Hg Conc. All Data (ng/L)	Avg Hg Conc Excl. outliers (ng/L)	Avg Hg Conc. NDs at MDL (ng/L)	Avg TSS Conc. (mg/L)
MCES							
Colby Lake	7	11/97, 4/98	NA	61	61	69	NA
Weir Drive	9	11/97, 4/98	NA	32	32	35	NA
Juliet St.	6	11/97, 4/98	NA	51	51	46	NA
Lilac-Men. Heights	5	6/98	NA	239	239	239	NA
Navajo-MH	6	6/98	NA	53	53	53	NA
So. St. Paul	6	6/98	NA	70	70	70	NA
MWRA							
DEDH	63	1/96 – 1/99	36	157	110	126	NA
WALT	63	1/96 – 1/99	26	149	110	139	NA
WEYM	63	1/96 – 1/99	47	102	102	122	NA
WINC	63	1/96 – 1/99	12-124	89	79	110	NA
HRSD							
Courthouse Estates	1	3/12/99	<10	17	17	17	143
Lago Mar	1	3/12/99	15	31	31	31	58
Hunt Club	1	3/12/99	15	86	86	86	245
Monroe Place	1	3/19/99	60	47	47	47	69
Elmhurst	1	3/19/99	50	284	284	284	176
Powhatan	1	3/19/99	50	58	58	58	120
Palo Alto							
Pulgas (mainly apartments)	45	5/95 – 2/99	37	292	172	192	NA
Waverly (upscale houses)	41	5/95 – 2/99	37	165	149	193	NA
Mean				178	138	143	
Median				110	104	88	
St. dev.				258²	112	223	

¹ NA – the data are not available.

² The results for the full data set indicated that the data significantly departed from normality including outlier concentrations. Therefore, any statistical evaluations regarding standard deviation (e.g., ANOVA) tend to be invalidated.

- There were seasonal differences in mercury concentrations in the sampled areas with higher concentrations observed during the second and third quarter of the calendar year. Application of an ANOVA test (99% confidence level) to the data sets that were considered to have a normal distribution determined that the seasonal trend was statistically significant.
- No clear correlation could be established between the age of the collection system/neighborhood and the mercury concentrations. Based on the Palo Alto data, population density may have a greater impact on mercury concentrations than the age of the service area.
- As can be seen from Table 2, a high level of variability was observed for the different locations sampled, with means for different locations varying from 17 to 284 ng/L and a number of non-detects and outliers. There are a number of reasons that this type of variability could occur as discussed in Sections 5.3 and 6.0 of the report.

3.0 PORTABLE TOILET WASTES

For comparison purposes, data were obtained and evaluated for portable toilet wastes, including chemical toilet wastes and septage. The intent of presenting these data was not for use in a mass balance, but to provide anecdotal support that levels of mercury in human excrement, independent of the contribution from discharged household products, are substantial.

3.1 Chemical Toilet Waste Results

Chemical toilet waste samples were collected and analyzed by the Northeast Ohio Regional Sewer District (NEORSDD), in Cleveland, Ohio. Because these wastes do not contain household products, the data collected should provide an indication of the fecal/urinary contribution of mercury to domestic wastewater. Eight samples of portable toilet wastes were collected in March and April 2000, along with a sample of the chemical treatment solution to evaluate its potential contribution to the toilet waste mercury concentrations. The results, which are shown in Table 3, yielded mean and median mercury concentrations of 3,737.5 ng/L and 800 ng/L. Although mercury was detected at 410 ng/L in the chemical treatment solution added to portable toilet waste, the solution volume is very small relative to the total waste volume, and cannot account for the total mercury measured in the waste.

Table 3. Summary of Mercury Concentrations in Portable Toilet Waste

Date	Detection Limit (ng/L)	Mercury Conc. (ng/L)	Assumed Mercury Conc. ng/L
3/17/00	1600	5800	5800
3/21/00	1600	ND	800
3/22/00	1600	3100	3100
3/29/00	1600	18000	18000
4/03/00	1600	ND	800
4/04/00	50	710	710
4/05/00	50	290	290
4/06/00	50	400	400
Mean			3737.5
Median			800

3.2 Septic Waste Results

NEORSD also collected and analyzed 34 samples from 12 different waste hauler services to determine concentrations of mercury in exclusively domestic septage. The results, which are presented in Table 4, yielded mean and median mercury concentrations of 12,918 ng/L and 6,950 ng/L.

Table 4. Summary of Mercury Concentrations in Septic Hauler Waste

Date	Detection Limit ng/L	Mercury Conc. ng/L	Assumed Mercury Conc. ng/L
2/15/00	200	5,400	5,400
2/16/00	200	3,900	3,900
2/16/00	200	2,700	2,700
2/16/00	1,600	1,300	13,000
2/21/00	1,600	1,900	1,900
2/21/00	1,600	6,700	6,700
2/22/00	1,600	4,100	4,100
2/22/00	1,600	17,000	17,000
2/23/00	1,600	7,800	7,800
2/23/00	1,600	4,600	4,600
2/24/00	1,600	11,000	11,000
2/24/00	1,600	6,000	6,000
2/25/00	1,600	120,000	120,000
2/24/00	1,600	9,400	9,400
2/28/00	1,600	7,200	7,200
2/29/00	1,600	6,000	6,000
2/29/00	1,600	2,600	2,600
3/01/00	1,600	5,100	5,100
3/01/00	1,600	13,000	13,000
3/02/00	200	19,000	19,000
3/02/00	200	11,000	11,000
3/07/00	1,600	26,000	26,000
3/07/00	1,600	4,600	4,600
3/08/00	1,600	7,700	7,700
3/08/00	1,600	1,700	1,700
3/11/00	1,600	5,100	5,100
3/11/00	1,600	ND	800
3/14/00	1,600	43,000	43,000
3/14/00	1,600	4,100	4,100
3/15/00	1,600	10,000	10,000
3/15/00	1,600	ND	800
3/15/00	1,600	35,000	35,000
3/16/00	1,600	11,000	11,000
3/16/00	1,600	12,000	12,000
Mean			12,918
Median			6,950

4.0 MERCURY IN HOUSEHOLD PRODUCTS

Some literature sources report commonly used detergents and toiletries as potentially contributing to mercury in residential wastewater. HRSD performed mercury determinations on several common household and toiletry items. The products analyzed were selected to represent those commonly used by the average consumer. The intent of these analyses was not to look at specific products, but to provide a snapshot of a group or category of products. To completely characterize a product, it would be necessary to analyze different brands, lot numbers, manufacturing facilities, etc., which was beyond the scope of this project. In addition, it was implicitly understood that a bias might result from analyzing the products only one time. Because these biases may be positive or negative, the average result of the biases were considered negligible to the final outcome of the calculations. The results of the analysis are presented in Table 5 according to product type.

Table 5. Mercury in Common Household Products and Toiletries

Product Type	Number Products Tested	Range of Concentration (ng/L)	
		Minimum	Maximum
Toothpaste	5	490	3,800
Shaving Cream	4	90	670
Deodorant/Antiperspirant	2	1,010	1,350
Soap/Shampoo	5	835	25,000
Toilet Tissue	3	220	1,510
Laundry Detergent	6	560	2,490
Bleach	2	<200	6,170
Dish/Dishwasher Detergent	4	560	1,320
Drain Cleaners	2	2,970	5,490
Soft Drinks/Drink Mixes ¹	3	25	6,070
Fruit Juices	3	789	3,560
Fruit/Vegetables ²	4	116	874
Rice/Grains	2	26	<200
Processed Meats	6	<100	290
Beef/Chicken	2	29	<40
Condiments ³	4	133	1,956
Food Coloring ⁴	4	96	137,000
¹ With yellow or red dyes. ² Fresh, frozen and canned. ³ Salt and sugar.			

5.0 MASS BALANCE FOR DOMESTIC SOURCES OF MERCURY

Mass balance calculations were performed using the mercury data for the domestic products evaluated to determine the relative contribution of these products to domestic mercury loadings to POTWs. In addition, mercury data for processed foods, meats, seafoods, vegetables, fruits, and seasonings were obtained and utilized in the calculations.

5.1 Assumptions

Because product and food consumption can vary widely, a number of assumptions were used in developing these calculations.

- The average household consists of four people (2 adults and 2 children).
- The average flow per household is 12,000 gal/month (45,420 L/month) (*Wastewater Engineering Treatment, Disposal, and Reuse Third Edition*, Metcalf & Eddy, 1991; *Sanitation Districts of Los Angeles County, Final Joint Outfall System Master Facilities Plan*, June 1995).
- Product use or consumption per month was determined using information from the American Dietetic Association (*ADA Food Pyramid and Website*), USDA recommended portions (*USDA Website*), and other estimates, which are presented in Appendix A.
- The contribution from fish and shellfish consumption is based on estimates cited in the USEPA *Mercury Study Report to Congress (EPA-452/R-97-006, Vol. IV, Tables 4-73 and 4-74)*. Children were estimated to have the same contribution rates as adults: a worst case scenario.
- Similar products (e.g., ground beef and chicken) with like concentrations were combined for the calculations.
- These analyses provide a snapshot of those products used by the average consumer. Samples selected represent only a cross-section of commonly used products.
- Since the entire product list was used in calculating the mass balance, it was understood that any biases would be negligible to the final result.
- Usage rates are *overall* averages and may vary from person to person.

5.2 Calculations

Mercury contributions from each product were calculated by multiplying the estimated quantity of product used per month by the average mercury concentration in the product. Using shaving cream as an example:

Usage	0.24 kg/month
Mercury Conc.	340 ng/kg
Mercury Contribution	= Usage x Conc.
	= 0.24 kg/month x 340 ng/kg
	= 81.6 ng/month

Fish and shellfish consumption calculations were based on information found in Tables 4-73 and 4-74, Volume IV of the *Mercury Study Report to Congress*. The 50th percentiles for total U.S. mercury exposure from fish and shellfish of marine, estuarine, and freshwater origin are 110 ng/kg/day and 100 ng/kg/day for men and women, respectively.

Exposure Men 110 ng/kg bw/day
 Exposure Women 100 ng/kg bw/day
 Weight Men 70 kg
 Weight Women 65 kg

Mercury Contribution_(M) = Exposure x Weight
 = 110 ng/kg bw/day x 70 kg x 30 day/month
 = 231,000 ng/month

Mercury Contribution_(W) = 100 ng/kg bw/day x 65 kg x 30 day/month
 = 195,000 ng/month

5.3 Results

Table 6 lists the products used in calculations, the estimated quantity of each product used per month, and the average mercury contribution from each source per household.

Table 6. Mass Balance for Domestic Sources of Mercury

Product	Usage per Month kg/month	Avg. Hg Conc. ng/kg	Hg Household Contribution ng/month
Shaving Cream	0.24	340	82
Deodorant	0.06	1180	71
Soap	0.12	7908	949
Shampoo	2.04	835	1,703
Toothpaste	0.42	1230	517
Mouthwash	0.91	15	14
Dishwashing Detergent	0.91	1320	1,201
Dishwasher Detergent	2.00	1478	2,956
Laundry Detergent	4.00	1478	5,912
Bleach	0.90	6170	5,553
Toilet Paper	1.00	827	827
Drain Cleaners	0.18	4230	761
Soft Drinks:			
Powdered Mix	0.14	6070	850
Premixed	5.52	25.1	139
Carbonated	8.16	142	1,159
Fruit Juice	14.7	2570	37,779
Rice/Grains	33.6	26.4	887
Hot Dogs/Sausage	3.64	100	364
Processed Lunch Meat	1.82	200	364
Fish and Shellfish ¹			852,000
Ground Beef, Chicken	7.23	30	217
Fresh, Frozen and Canned Vegetable and Fruit	27.6	400	11,040
Sugar ²	6.00	1602	9,612

Product	Usage per Month kg/month	Avg. Hg Conc. ng/kg	Hg Household Contribution ng/month
Salt ²	1.20	1,956	2,347
Total All Products			937,303
¹ Monthly consumption in a four person household.			
² Includes quantities found in prepared foods.			

To determine the average domestic contribution per household, the sum of the mercury contribution for all of the domestic and food products was divided by the flow for each household.

Total Mercury Contribution 937,303 ng/month
Average Household Flow 45,420 L/month

Average Household Contribution = Mercury Contribution ÷ Average Flow
= 937,303 ng Hg/month ÷ 45,420 L/month
= **20.6 ng/L**

Based on the estimated average household mercury contribution of 20.6 ng/L and the mean mercury domestic wastewater concentration of 138 ng/L (data set excluding outliers), approximately **15 % of the total domestic contribution can be attributed to food, toiletry and household products**. With this information, it is apparent that at least **85% of the mercury in domestic waste comes from other sources**.

One explanation is that a significant source of mercury comes from human wastes. Information in the literature indicates that after mercury is released from human tissues, fecal excretion becomes the predominant route for elimination of mercury from the body, and that the rate of excretion correlates with the number of amalgam fillings (*Lorscheider, et al., Mercury Exposure From Silver Tooth Fillings: Emerging Evidence Questions a Traditional Dental Paradigm, The FASEB Journal, 9: 504-508, 1995*). Engqvist has shown that the amalgam particles are formed when a person chews aggressively, and the amount of particles originating from fillings can be estimated from a fecal sample, with at least 80% of the ingested particles excreted (*Engqvist et al., Speciation of Mercury Excreted in Feces from Individuals With Amalgam Fillings, Archives of Environmental Health, 53: 205-213, May/June 1998*). This same study showed that mercury vapor dissolved in water and swallowed was only excreted to about 40% in feces.

Measurements by Skare (*Water, Air, and Soil Pollution 80: 59-67, 1995*; see Appendix B) have indicated that dental amalgam-loaded individuals excrete mercury at average rates of 64 ug/day in feces and 4.5 ug/day in urine. The individuals studied were selected to represent a broad range in amalgam loadings, and none normally ate fish from lakes or smoked. The control from the Skare study, representing dental amalgam-free individuals, had measured mercury excretion rates of 1 ug/day in feces and 0.4 ug/day in urine. Subtracting the control measurements, the total rate of dental amalgam mercury excreted by the average dental amalgam-loaded individual through feces and urine would be 67 ug/day (67,000 ng/day).

In the *Mercury Study Report to Congress* it was reported that the individuals with dental amalgams are exposed to elemental mercury vapor released by these fillings. Mercury vapors are almost entirely re-absorbed by the lungs and eventually excreted in urine at the rate of 1-5ug/day (*Vol. IV, 5-1*). The 1-5 ug/day "general" range presented in represents urinary excretion only, and is not inconsistent with the 1995 Skare study.

It should also be noted that some mercury is excreted in hair. In Table 6-3, Volume IV, of the *Mercury Study Report to Congress*, the average mercury concentration in hair from studies in various U.S. communities is approximately 1 to 2 ug/g. It is difficult to estimate the average growth rate of hair in order to calculate the average rate at which mercury is excreted through hair. However, this source is believed to be quite small².

Assuming a daily wastewater generation rate of 100 gallons per day per individual (378 L/day; *Sanitation Districts of Los Angeles County, Final Joint Outfall System Master Facilities Plan, June 1995*) and an estimate that 65% of the population has dental amalgam fillings (*September 1992 Bio-Probe*), a domestic sanitary sewage mercury concentration attributable to excreted dental amalgam mercury can be predicted from the Skare data as follows:

Rate of mercury excretion	67,000 ng/day
Water usage	378 L/day
Percentage of amalgam population	65%
Mercury conc. from excreted wastes	= 0.65 x 67,000ng/day ÷ 378 L/day
	= 115.2 ng/L

Using the mean mercury domestic wastewater concentration of 138 ng/L, this ***loading from feces and urine could account for 83 % of the total domestic loading.*** Considering the variability in domestic waste concentrations and uncertainties in the percentage of the population with amalgam fillings, this percentage could be higher or lower than this estimate, but this source of mercury is certainly significant.

Applying the Skare data to the NEORS D service area population of 1.1 million, the human-excreted dental amalgam mercury loading to the NEORS D sewerage system can be estimated:

Amalgam loading	= populations affected x excretion rate
	= 1,100,000 x 0.65 x 67,000 ng/day ÷ 10⁹ ng/gram
	= 48 grams/day

Based on the average NEORS D total treated flow of one billion L/day and a treatment plant mercury removal efficiency of 97 percent, ***48 grams/day of human-excreted dental amalgam mercury is, by itself, enough mercury to exceed the 1.3 ng/L water quality criterion*** at the NEORS D plant effluents:

Effluent concentration	= removal x loading ÷ flow
	= (1 - 0.97) x 48 grams/day x 10⁹ ng/gram ÷ 10⁹ L/day
	= 1.4 ng/L

Due to the limited number of individuals studied by Skare as well as uncertainty about how representative these individuals may be, some questions remain concerning numeric values derived from the Skare study data. Nonetheless, the Skare study results indisputably indicate that the contribution to domestic wastewater from excreted dental amalgam mercury is substantial. At the very least, these results demonstrate that further consideration of this mercury source is imperative.

² For comparison purposes, to equal the estimated 67 microgram/day rate through feces and urine excretion, hair would have to grow at a rate of 45 grams/day or 3 pounds/month.

Another potential source of mercury in domestic wastewater is the improper disposal of thermometers. A study performed by Larry Walker Associates (*Davis, CA 1994*) estimated that on an annual basis, 1.6% of households discarded 1 thermometer to the sewer, with each thermometer contributing 0.5 g. Using our original assumption of 4 individuals per household, the NEORSD service area could be contributing 6 grams/day based on improper disposal of thermometers.

Thermometer disposal	=	discard rate x households x therm. contrib.
	=	0.016 x 275,000 households x 1 therm/household/yr
		x 0.5 gram/thermometer ÷ 365 days/year
	=	6 grams/day

Other potential sources could include infiltration and inflow from rain or rising groundwater, the drinking water sources, ritualistic uses and vaccinations for children.

Infiltration and inflow contributions will vary depending on location, geology¹ and climate. Indeed, the impacts from infiltration and inflow may explain why the domestic mercury wastewater data showed statistically significant differences in concentrations. This observation has been validated by other POTWs, including biosolids data provided by the Western Lake Superior Sanitary District in Duluth, Minnesota. The data show lower mercury concentrations in the winter when the ground is frozen, in comparison to spring and summer conditions (*Personal communication, data collected August 1985 – February 2000, Tim Tuominen*).

Some limited data were available on the levels of mercury in the tributary drinking water sources for the agencies studied using sensitive mercury methods. For the HRSD service area, five tap water samples were collected to characterize the five treatment facilities in the service area. The average concentration was 0.7 ng/L. In the Minneapolis area, 8 samples were collected from the service area in March 2000 with an average mercury concentration of 0.4 ng/L. It should be noted that in order fully characterize drinking water from these areas, it would be necessary to conduct multiple sampling capturing seasonal changes. The City of Palo Alto collected 12 water samples during the period March 1999 - April 2000 with an average mercury concentration of 0.9 ng/L. These data confirm other information that shows that drinking water is a relatively minor contribution to the POTW mercury loadings. The range in concentration depends on whether the source is surface water (3 - 4 ng/L for some Great Lakes communities; *Personal communication, Keith Linn, NEORSD*) to groundwater (2 ng/L) for Wisconsin. In these cases, the relative contribution from drinking water to total wastewater mercury appears to be small (2 to 4%)

Studies conducted by the Chicago Department of Public Health have shown that mercury is used in religious rituals and folk medicines in Chicago's Hispanic community (*Potential Dangers of Using Mercury in Religious Rituals and Folk Medicines, US EPA Region 5, September 1997*). Ritual mercury users obtain mercury from *botánicas*, friends and folk healers.

Thimerosal is a mercury-containing preservative that has been used as an additive to biologics and vaccines since the 1930's to kill bacteria used in vaccines and in preventing bacterial contamination.

¹ The average abundances for common geological materials are: (I) igneous rocks—0.004 mg/kg, ultramafic; 0.01 mg/kg, mafic; 0.04 mg/kg, granitic; (ii) sedimentary rocks---0.04 mg/kg, limestone; 0.03 mg/kg, sandstone; 0.02 to 0.40 mg/kg, shale; (iii) soils---0.056 mg/kg. Rose, J.A., H.E. Hawkes, and J.S. Webb. 1979. *Geochemistry in mineral exploration*. 2nd ed. Acad. Press, New York.

Some, but not all of the vaccines recommended routinely for children in the U.S. contain thimerosal. The U.S. Public Health Service and the American Academy of Pediatrics are working collaboratively to assure that the replacement of thimerosal-containing vaccines takes place as expeditiously as possible (*Centers for Disease Control and Prevention Website, Bulletins: Thimerosal, May 15, 2000*). Manufacturers have been asked for a clear commitment and a plan to eliminate or reduce the mercury content of their vaccines.

6.0 CONCLUSIONS

The results of this study offer some important observations for sources of mercury in domestic wastewater and the feasibility of effective control options.

- Significant amounts of mercury at the average concentration of 138 ng/L were consistently found in strictly domestic wastewater in various parts of the country.
- An important finding in the statistical data evaluation is that regardless of the approach used (excluding outliers and including non-detectable values) the mean mercury concentrations were not significantly affected.
- A correlation between the age of the neighborhood and the mercury concentrations was not substantively meaningful. It appears that the more significant factor may be the density of population in the neighborhoods. Unfortunately, only very limited information regarding the population density was available.
- Statistically significant differences were found when the data were grouped by quarters. The mercury concentrations in residential wastewater appear to be higher in second and third quarter of the calendar year. This can potentially be explained as impacts resulting from infiltration and inflow to sewers during rainy seasons.
- The variability in the levels of mercury observed in the domestic wastewater samples may also be the result of differences in the number of amalgam surfaces per individual, fish/shellfish consumption rates, water usage, water source, and rates of mercury settling/resuspension in sewers.
- Several common household and toiletry items were found to contain substantial concentrations of mercury when examined using sensitive analytical techniques. Although these products individually do not contribute a lot to a total concentration in wastewaters, their cumulative effect accounts for approximately 15% of the mercury concentration in domestic wastewater. The feasibility of controlling these sources would require a national effort.
- Although several sources contributing to the domestic mercury concentrations have been identified, human wastes (feces and urine) from amalgam loaded individuals are believed to be the most significant source (> 80%).
- These results were corroborated by the results from the chemical toilet and septic wastes that showed that a significant portion of the mercury in domestic wastewater is from uncontrollable sources such as dental amalgam fillings.
- While controlling human wastes is impractical, the long-term outlook is promising inasmuch as

the trend in dental health is for fewer cavities and resulting in smaller and smaller populations of amalgam-loaded individuals over time.

Based on this information, domestic waste contributes appreciable concentrations of mercury to POTW influent wastestreams and must be considered when addressing mercury control strategies and the likelihood of virtual elimination of mercury. Background mercury concentrations averaging more than 100 ng/L can be expected in POTW wastewater influents, even if complete elimination of industrial point source discharges is accomplished.

In EPA's cost analysis for the Great Lakes Water Quality Initiative, and in subsequent discussions with wastewater representatives, the Agency has supported the use of pollutant minimization programs as a way for achieving compliance. AMSA strongly endorses and promotes pollution minimization efforts, but is concerned that these efforts may not be adequate to produce the desired level of permit compliance sought by regulatory authorities, highlighting the need for a national compliance strategy for POTWs.

APPENDIX A

Assumptions Used to Determine Usage for Consumer Products

The assumptions used to determine usage quantities per month for mass balance of household products, toiletries and food items found in Table 5 are detailed below. The number of events per month are based on a four family member household, such that:

$$\begin{aligned} \text{Number events per month} &= 4 \text{ people} \times 1 \text{ event/person} \times 30 \text{ days/month} \\ &= 120 \text{ events/month} \end{aligned}$$

Usage was determined using specific quantities used per person per event or based on the amount of product routinely purchased per month. The monthly usage was calculated as follows:

$$\begin{aligned} \text{Monthly usage} &= \text{Usage per event} \times \text{Number of Events} \\ &= \text{Usage per month} \end{aligned}$$

The following are specific items that were used in the mass balance calculation:

Shaving Cream

Usage/Event	0.002 kg
No. of Events	120
Usage/Month	0.24 kg

Deodorant

Usage/Event	0.0005 kg
No. of Events	120
Usage/Month	0.06 kg

Soap

Usage/Event	0.001 kg
No. of Events	120
Usage/Month	0.12 kg

Shampoo

72 oz/month	
Usage/Month	2.04 kg

Toothpaste

15 oz/month	
Usage/Month	0.42 kg

Mouthwash

32 oz/month	
Usage/month	0.91 kg

Dishwashing Detergent

32 oz/month	
Usage/Month	0.91 kg

Dishwasher Detergent

Usage/Event	0.10 kg
No. of Events	20
Usage/Month	2.00 kg

Laundry Detergent

Usage/Event	0.10 kg
No. of Events	40
Usage/Month	4.00 kg

Bleach

32 oz/month	
Usage/Month	0.90 kg

Toilet Paper

4 Rolls/month (0.25 kg/roll)	
Usage/Month	1.00 kg

Drain Cleaners

36 oz/month	
Usage/Month	1.00 kg

Soft Drinks

Powdered Mix	
Usage/Event	0.007 kg (~0.25 oz. pack)
No. of Events	20 packs/month
Usage/Month	0.14 kg

Premixed	
Usage/Event	0.23 kg (~8 oz. bottle)
No. of Events	24 bottles/month
Usage/Month	5.52 kg

Carbonated Drinks

Usage/Event	0.34 kg (12 oz. Can)
No. of Events	24 cans/month
Usage/Month	8.16 kg

Fruit Juice

Usage/Event	0.12 kg (~4 oz. Serving)
No. of Events	120
Usage/Month	14.7 kg

Rice/Grains

Usage/Event	0.28 kg (~10 oz. Serving)
No. of Events	120
Usage/Month	33.6 kg

Hot Dogs/Sausage

Usage/Event	0.11 kg (~4 oz. Serving)
No. of Events	32
Usage/Month	3.64 kg

Processed Lunch Meat

Usage/Event	0.11 kg (~4 oz. Serving)
No. of Events	16
Usage/Month	1.82 kg

Ground Beef Chicken

Usage/Event	0.11 kg (~4 oz. Serving)
No. of Events	64
Usage/Month	7.23 kg

Fresh, Frozen, and Canned Fruits and Vegetables

Usage/Event	0.23 kg (~8 oz. Serving)
No. of Events	120
Usage/Month	27.6 kg

Sugar

Usage/Event	0.01 kg (~4 teaspoons)
No. of Events	600
Usage/Month	6.00 kg

Salt

Usage/Event	0.01 kg (per ADA website)
No. of Events	120
Usage/Month	1.20 kg

APPENDIX B

Mass Balance and Systemic Uptake of Mercury Released From Dental Amalgam Fillings

I. Skare

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MASS BALANCE AND SYSTEMIC UPTAKE OF MERCURY RELEASED FROM DENTAL AMALGAM FILLINGS

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Abstract. The release of mercury (Hg) from dental amalgam fillings has been verified by several authors. In this study, the emission rate of Hg⁰-vapor from the oral cavity (O-Hg) and the urinary Hg-excretion rate (U-Hg) have been studied with 34 healthy individuals. In ten cases, the urinary excretions of silver (U-Ag) and the fecal excretions of Hg and Ag (F-Hg, F-Ag) were also monitored. All variables, except U-Ag, were significantly related to the load of amalgam. According to this study, an individual with a moderate load of amalgam, i.e. 30 restored surfaces, is predicted to exhibit the following emission rates: O-Hg=22, U-Hg=3, F-Hg=60 and F-Ag=27 µg/d (d=24 hours), consistent with a gross mass balance for Hg of approximately 60 µg/d. The corresponding systemic uptake of Hg was estimated to 12 µg/d based on external data relating air Hg⁰-exposures to urinary Hg-excretions. The worst case individual showed a gross mass balance of 200 µg Hg/d connected to a systemic uptake of 70 µg Hg/d. These values were compared to the average intake of total-Hg by a Swedish diet (2 µg/d) and to the WHO's tolerable value for intake of total-Hg by food (45 µg/d). Upscaled to the entire Swedish population (8 mill.), the data suggests a fecal/urinary emission to the environment of 100 kg Hg yearly originating from a population load of amalgam fillings containing 90,000 kg of Hg.

1. Introduction

Before the beginning of the 1980's, the stability of dental amalgam with respect to the release of mercury (Hg) was generally not very much questioned. The release of Hg from amalgam has, however, since then been described and verified by several authors (Aronsson, 1989; Berglund, 1988, 1990; Björkman, 1992; Brune, 1985; Frykholm, 1957; Jokstad, 1992; Patterson, 1985; Pleva, 1992; Skare, 1994; Svare, 1981; WHO, 1991; Vimy, 1985a, 1985b).

For individuals with a moderate load of amalgam, i.e. approximately 30 restored surfaces, a basic release rate of elemental Hg⁰ from the oral cavity of 20 µg/d (d=24 hours) is normally averaged, reaching about 100 µg Hg⁰/d for such individuals most heavily loaded. By chewing and by drinking hot beverages the Hg⁰-emission may temporarily be increased by three to tenfold.

One part of the released elemental Hg⁰ is exhaled, and one part is retained in the saliva and swallowed together with amalgam particles and corrosion products, giving a gastrointestinal inorganic-Hg flow from which, however, only a smaller fraction is supposed to be systemically absorbed (WHO, 1991).

The remaining part of the released Hg⁰ should be systemically absorbed through the lungs or by resorption through the oral mucosa. Being uncharged and monoatomic, elemental Hg⁰ is a highly mobile species capable of entering most of the body compartments. The systemic uptake of Hg is, in addition to the present number of amalgam fillings, also influenced by the mean ratio of the oral-to-nasal breathing and to the actual chewing pattern.

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For risk assessment of a long-term inorganic-Hg exposure, biological monitoring of the urinary Hg-excretion is normally applied. Several authors have significantly related elevated urinary Hg-levels to the load of amalgam (Aronasson, 1989; Berglund, 1990; Jokstad, 1992; Langworth, 1991; Olstad, 1987; Skare, 1990, 1994; WHO, 1991; Åkesson, 1991).

Other common biological monitoring indices used are the total-Hg plasma concentration and the level of inorganic-Hg in whole blood - designed for minimizing the confounding influence from MeHg present in the blood. Some authors have used the blood matrix when studying the Hg-exposure from amalgam (Jokstad, 1992; Langworth, 1991; Snapp, 1989; Svare, 1981; Åkesson, 1991).- Only few studies have been published where the monitoring of fecal Hg-excretion is attentioned (Frykholm, 1957; Stock, 1934).

In the present study, *the gross mass flow balance of Hg and the systemic uptake of Hg* have been estimated for individuals with different load of amalgam fillings, based on data from the monitoring of oral air Hg-emissions and excretion rates of Hg/Ag by urine and feces. In addition, the Hg-excretion data has been upscaled to represent the entire Swedish population, thus obtaining an estimate of the Hg-emission from amalgam fillings to the sewage systems and to the environment.

2. Subjects and methods

2.1. TEST SUBJECTS AND MEASUREMENTS

Basic monitoring of the emissions of oral air Hg and urinary Hg were performed with 34 healthy adult individuals of both sexes, occupationally unexposed to Hg or Ag. In ten cases, the urinary excretions of silver (U-Ag) and the fecal excretions of Hg and Ag (F-Hg, F-Ag) were also monitored. The individuals were selected to represent a broad range in amalgam loading. None of them normally eat fish from lakes and none was a present smoker. The number of restored amalgam surfaces (N) was examined by a dentist.

2.2. ANALYTICAL METHODS

2.2.1. Elemental Mercury Vapor Emission into the Oral Cavity

As amalgam surfaces are very easily influenced by all kinds of mechanical and chemical actions, the oral environment should be properly normalized prior sampling. Two different methods for monitoring of the emission of oral air Hg⁰ have been used in the study (Skare, 1994).

By one method, a well-defined flow of oral air (1.5 l/min) was continuously sampled through a mouth-piece and passed into a gas cell of a UV-instrument until a steady-state reading was established, from which the oral emission rate of Hg⁰ was calculated. The lowest quantifiable Hg⁰ vapor concentration in the cell, i.e. 1 µg Hg⁰/m³, corresponds to a oral Hg⁰ emission rate of approximately 2 µg/d (d=24 hours).

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The other oral air Hg⁰-method was based on the covering of all amalgam surfaces with a 25-ml portion of water for a fixed period of time (2.0 min) by that collecting all Hg⁰-vapor emitted into the aqueous phase. The content of total-Hg in the aqueous sample was determined, after a wet-digestion step at room temperature with an acidified potassium permanganate (KMnO₄) solution, by using a standard cold AAS-technique (D.L.= 0.2 ng Hg/ml) implying a releasing step to Hg⁰-vapor by Sn(II)-reduction (Skare, 1972).

The detection limits, equal for the two oral air Hg⁰-methods, correspond to a Hg⁰-rate expected from 2-3 restored amalgam surfaces, i.e. 1-2 µg O-Hg/d.

2.2.2. Total Mercury and Silver Excretions by Urine

The individuals were requested to collect all their urine voided during a 24-hour period. Sub-samples were wet-digested at room temperature by the addition of an acidified KMnO₄-solution.

The total content of U-Hg was determined by using the standard cold AAS- technique. The detection limit, expressed as excretion rate, was on the average 0.2 µg U-Hg/d.

The total content of U-Ag was determined using a graphite furnace AAS- technique. The detection limit, expressed as excretion rate, was on the average 0.6 µg U-Ag/d.

2.2.3. Total Mercury and Silver Excretions by Feces

The individuals were requested to deliver two consecutive fecal voidings while recording the duration intervals. The contents of Hg and Ag in the samples were, after digestion with warm concentrated HNO₃, determined by using an ICP-technique (external laboratory). After relating the results to the sampling times, the results were converted to excretion rates for Hg and Ag (Skare, 1994).

The lowest quantifiable excretion rates were approximately 3 µg F-Hg/d and 0.8 µg F-Ag/d, respectively.

2.3. MASS BALANCE CALCULATIONS ON MERCURY

In estimating the total exposure from Hg, i.e. the gross balance of Hg passing the body, for individuals occupationally unexposed to Hg, the following Hg-containing sources should initially be considered: air, food and water supply, and amalgam fillings.

Urban air may contain 5 ng Hg⁰/m³. If the mean breathing rate during the day is limited to 15 l/min and an uptake efficiency of 80% is assumed, then, the contribution to the systemic Hg-uptake by breathing environmental air may not exceed 0.1 µg Hg/d.

The mean contribution of total-Hg from a Swedish dietary (for normal fish consumers) has recently been determined to approximately 2 µg Hg/d, about 2/3 of which are present as MeHg species (Becker, 1991). The systemic uptake of Hg from the intestines is considered to be 90% for MeHg and 5 to 10% for inorganic Hg-species (WHO, 1991).

The purpose of monitoring the fecal Ag-excretion was the possibility to make an indirect estimation of the fraction of Hg contained in the released and swallowed particles and corrosion species. At time of insertion, amalgam restorations contain Hg and Ag in a ratio of approximately 1:0.7 by weight. If the composition of the surface layers is

assumed to be constant for years, a rough estimate of Hg contained in particles and corrosion products can be calculated from the F-Ag rate after correction for food-Ag in feces (obtained from the F-Ag excretion with the amalgam-free individual).

The estimation of the systemic uptake of Hg is a more elusive task. However, since the systemic input and output of Hg, at equilibrium, by definition is the same, the systemic uptake of Hg should be more adequately estimated by using urinary excretion data than by using data related to intake patterns.

After entering the blood, most of the body-burden of Hg (90%) is stored in the kidneys, the Hg-content of which is reflected by the U-Hg excretions. A minor part of the systemic-Hg is expected to be stored in other tissues exhibiting very long half-times for Hg-clearance (i.e. years), where the equilibrium is very slowly attained. This latter fraction is, however, not predictable from urinary data (WHO, 1991).

In equilibrium with a long-term inorganic Hg-exposure, the daily urinary Hg-excretion has been shown to be rather constant (Skare, 1994). By mathematical integration of the kidney-clearance decay curve to infinity, assuming $t_{1/2} = 45$ days, a first order of kinetics and by using the monitored U-Hg rate as input data, the kidney-burden can be estimated by the equation:

$$\text{Kidney-burden } (\mu\text{g Hg}) = \text{U-Hg}(\mu\text{g/d}) \cdot 45(\text{d}) / \ln 2.$$

For example, an individual with a moderate load of amalgam, exhibiting at equilibrium a daily urinary excretion of $3 \mu\text{g Hg}$ should have a kidney-burden of approximately $200 \mu\text{g Hg}$. Individuals, very heavily loaded with amalgam, may in extreme cases show urinary Hg-excretions (and kidney-burdens), which are tenfold higher.

The body-clearance of Hg is not accomplished only by urine but also by feces, sweat, exhalation and by storage in hair and nails. However, only the excretions by urine and feces are large enough to be considered. The fecal excretions contain Hg-species (maybe biotransformed), which partly have been swallowed and partly originate from Hg systemically absorbed and excreted through the bile.

To estimate the excretion rate of Hg through the bile, again, U-Hg excretion rate data might be helpful. Referring to the report by WHO 1991, an occupational air concentration of $25 \mu\text{g Hg}^0/\text{m}^3$ is, on group level, consistent with a urinary excretion of $45 \mu\text{g U-Hg/d}$ ($\approx 30 \mu\text{g U-Hg/g creatinine}$). As a daily 8-h Hg^0 -exposure is consistent with a systemic uptake of $175 \mu\text{g Hg/d}$ [calculation: $25(\mu\text{g}/\text{m}^3) \cdot 18(\text{l}/\text{min (worker)}) \cdot 8(\text{h}) \cdot 60(\text{min}/\text{d}) \cdot 80\%(\text{retention efficiency})$], the difference between this total-Hg uptake rate and the U-Hg rate ($45 \mu\text{g/d}$) should be the averaged bile-Hg rate.

If this relationship between the urinary and the bile Hg-excretion is also valid at lower levels of exposure, then an equation can be made up for estimating the total systemic uptake of Hg for individuals loaded with amalgam:

$$\Sigma \text{Hg}_{\text{uptake}} (\mu\text{g/d}) = 4 \cdot \text{U-Hg} (\mu\text{g/d})$$

This formula is not inconsistent with results from human Hg^0 -exposure studies reported by Cherian, 1978.

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3. Results and discussion

3.1. BIOLOGICAL MONITORING OF MERCURY AND SILVER

The results obtained from the ten individuals concerning oral air Hg⁰-emissions and urinary and fecal excretions of Hg and Ag are summarized in Table I.

As seen from Table I, the flow rates of Hg/Ag for the amalgam-free individual were, with one exception, i.e. U-Ag, very low compared to the corresponding rates for the nine amalgam loaded individuals. In spite of this group of nine individuals, on average, was somewhat heavier loaded with amalgam (mean: N=40 surfaces) than is normally expected from a Swedish middle-age group (i.e. N=30), the results still clearly indicate that man's exposures to total Hg and Ag predominately originate from the presence of dental amalgam fillings.

TABLE I
Emission and excretion rates of Hg and Ag from individuals loaded with amalgam fillings
(Data from ten individuals; Hg = mercury, Ag = silver and d = 24 hours)

Variable	Symbol	Amalgam loaded individuals		Control individual
		Md-value	Range	
Number of amalgam surfaces	N	40	18 - 82	0
Oral air Hg ⁰ emission	O-Hg	29 µg/d	20 - 124 µg/d	0 µg/d
Urinary Hg excretion	U-Hg	4.5 µg/d	1.8 - 19 µg/d	0.4 µg/d
Urinary Ag excretion	U-Ag	1.7 µg/d	1.4 - 6.0 µg/d	1.3 µg/d
Fecal Hg excretion	F-Hg	64 µg/d	27 - 190 µg/d	1 µg/d *
Fecal Ag excretion	F-Ag	33 µg/d	11 - 97 µg/d	4 µg/d *

* Mean value based on a homogenized sample from ten consecutive days

In this study, the emission rate of Hg⁰-vapor from the oral cavity has been determined by using two entirely different methods. The close accordance in the results by the two methods gives support for assuming the averaged O-Hg rates to be reliable. The method, in which only a simple water trap is used for sampling, might, because of simplicity, be the method choice for out-of-laboratory purposes. This does not say that the determination of the unstimulated O-Hg rate should be the most appropriate way in assessing an amalgam Hg-exposure.

According to several studies, urinary excretions do not often exceed 1 µg Hg/d for amalgam-free individuals occupationally unexposed to Hg. Our control individual did apply to this prediction. Urinary excretions, due to amalgam, exceeding 15 µg Hg/d are also rare. Our worst case individual, having 82 restored amalgam surfaces, many of which in a bad condition, showed a urinary excretion of 19 µg Hg/d.

The content of Hg in feces was about twice the content of Ag in feces. The extremely high coefficient of correlation for F-Hg vs F-Ag (Table II) should be a strong evidence for the fecal Hg-excretions being connected to the bearing of dental silver-amalgam. For individuals with a moderate load of amalgam (N=30 surfaces), their fecal Hg-excretion rate was predicted to be about 20 times the urinary Hg-excretion rate and about 30 times the total-Hg intake by food (2 µg Hg/d) consuming an average Swedish diet. This food-Hg value, reported by Becker and Kumpulainen 1991, was consistent with the fecal Hg-excretion value exhibited by the amalgam-free control individual.

TABLE II

Correlation coefficients (Pearson, *r*) among N, O-Hg, U-Hg, U-Ag, F-Hg and F-Ag
See Table I for explanation of symbols. The number of observations (*n*) used and the adherent statistical *p*-values are also displayed

	N	O-Hg	U-Hg	U-Ag	F-Hg	F-Ag
O-Hg	<i>r</i> = 0.82 <i>p</i> < 0.0001 <i>n</i> = 34	--				
U-Hg	<i>r</i> = 0.80 <i>p</i> < 0.0001 <i>n</i> = 34	<i>r</i> = 0.91 <i>p</i> < 0.0001 <i>n</i> = 34	--			
U-Ag	<i>r</i> = - 0.20 <i>p</i> = 0.59 <i>n</i> = 10	<i>r</i> = - 0.23 <i>p</i> = 0.53 <i>n</i> = 10	<i>r</i> = - 0.23 <i>p</i> = 0.53 <i>n</i> = 10	--		
F-Hg	<i>r</i> = 0.67 <i>p</i> = 0.033 <i>n</i> = 10	<i>r</i> = 0.85 <i>p</i> = 0.001 <i>n</i> = 10	<i>r</i> = 0.81 <i>p</i> = 0.003 <i>n</i> = 10	<i>r</i> = 0.12 <i>p</i> = 0.75 <i>n</i> = 10	--	
F-Ag	<i>r</i> = 0.74 <i>p</i> = 0.02 <i>n</i> = 9	<i>r</i> = 0.93 <i>p</i> < 0.0001 <i>n</i> = 9	<i>r</i> = 0.90 <i>p</i> = 0.0003 <i>n</i> = 9	<i>r</i> = 0.16 <i>p</i> = 0.69 <i>n</i> = 9	<i>r</i> = 0.97 <i>p</i> < 0.0001 <i>n</i> = 9	

Referring to Table II, significant levels of interplay were, excl. U-Ag, seen among all the emission/excretion variables and the number of amalgam surfaces. The deviating behavior of the U-Ag variable, e.g. no significant correlation to N, indicates that Ag from amalgam is not, or only to a very low extent, systemically absorbed.

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The following linear regression line equations have been calculated:

$$\text{O-Hg} = 0.1 + 0.73 \cdot N; \quad (\text{based on 34 observations})$$

$$\text{U-Hg} = 0.4 + 0.08 \cdot N; \quad (\text{based on 34 observations})$$

$$\text{F-Hg} = 15 + 1.45 \cdot N; \quad (\text{based on 10 observations})$$

$$\text{F-Ag} = 4 + 0.77 \cdot N; \quad (\text{based on 9 observations})$$

3.2. MASS BALANCE CALCULATIONS ON MERCURY

3.2.1. Gross mass balance of Hg

After a long-term Hg-exposure, the input and output flows of Hg should, at equilibrium equalize. The two main pathways for man's excretion of inorganic Hg are through urine and feces. Thus, a satisfactory estimate of the mean daily input of total-Hg should be the sum of the daily U-Hg and the F-Hg excretion rates. With a load of amalgam scored to be 30 restored surfaces, i.e. the average load of amalgam for middle-age people in Sweden, the predicted excretion rates (see regression equations above) should make up a gross mass balance being approximately 60 $\mu\text{g Hg/d}$.

In Figure 1, the intake, uptake and excretion flows of Hg for this average individual are outlined. As seen from Figure 1, the intake of amalgam-Hg is the most dominant origin to the Hg-exposure.

The elimination of all amalgam fillings should result in a very rapid decay of the F-Hg rate level. The U-Hg rate and some part of the bile-Hg rate, both reflecting the body-burden of Hg, should decline more slowly, i.e. during months.

The fecal Hg-excretions may consist of Hg-species as amalgam particles, corrosion products (oxidized Hg), bile-Hg (probably Hg connected to SH-groups in low-molecular weight proteins), biotransformed MeHg from food (mineralized by bacterial action) and species originating from the swallowing of elemental Hg^0 -vapor with the saliva. Some of these Hg-species may have passed the gastro-intestinal tract without any interactions at all, whereas other Hg-species have had a systemic past.

Upscaled to the entire Swedish population (8.5 mill.), the fecal-urinary excretions contain about 100 kg Hg/year originating from a population load of dental amalgam restorations containing approximately 90,000 kg of Hg.

3.2.2. Systemic uptake of Hg

The systemic uptake for an individual moderate loaded with amalgam ($N=30$ surfaces) has here been calculated to 12 $\mu\text{g Hg/d}$ based on urinary excretion data and assumptions concerning the relationship between air- Hg^0 exposure and U-Hg data (see 2.3). This amalgam Hg-exposure should be equivalent to a daily 8-hour occupational Hg^0 -exposure of 2 $\mu\text{g Hg}^0/\text{m}^3$, and corresponds to a total body-burden of 200 to 250 $\mu\text{g Hg}$.

Our worst case individual was suggested to exhibit a systemic uptake of 70 $\mu\text{g Hg/d}$, which value might be compared to a food standard by WHO stating the daily "tolerable" intake of total-Hg and MeHg should not exceed 45 and 30 $\mu\text{g Hg/d}$, respectively (WHO, 1972). The "acceptable" intake should be *none* according to the same report.

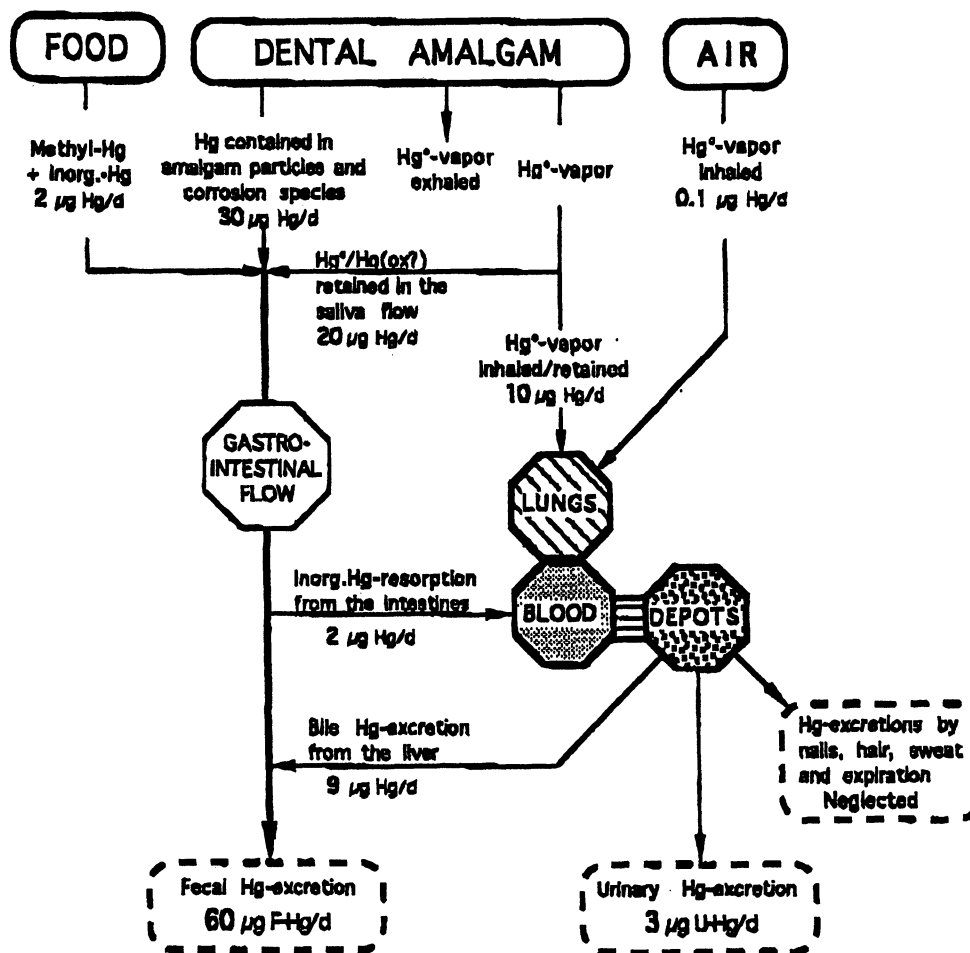


Figure 1. Flow chart exhibiting the intake, uptake and excretions of Hg originating from environmental air, food and dental amalgam.. Applicable to individuals with a moderate load of amalgam, i.e. 30 surfaces.

(For lucidity, the enterohepatic recirculation of MeHg is not outlined)

4. Conclusions

The study has confirmed that human emissions of oral air Hg⁰ and excretions of urinary Hg are significantly related to the present load of dental amalgam fillings. In addition, even the fecal excretions of Hg and Ag were shown to exhibit a significant relationship to the number of amalgam fillings.

For an individual with a moderate load of amalgam, the predominating part of the gross mass balance of Hg originates from the fillings. At comparison, a normal contribution to the Hg-exposure from air, water and food should be neglectable.

The daily systemic absorption of Hg was, for individuals heavily loaded with amalgam, predicted to be close to or even exceeding the WHO's recommendation for "tolerable" intake of total-Hg by food.

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References

- Aronsson, A. M., Lind, B., Nylander, M. and Nordberg, M.: 1989, *Biol Metals* 2, 25-30.
- Becker, W. and Kumpulainen, J.: 1991, *British J of Nutrition* 66, 1-9.
- Berglund, A., Pohl, L., Olsson, S. and Bergman, M.: 1988, *J Dent Res* 67, 1235-1242.
- Berglund, A.: 1990, *J Dent Res* 69, 1646-1651.
- Björkman, L. and Lind, B.: 1992, *Scand J Dent Res* 100, 354-360.
- Brune, D. and Evje, D.: 1985, *Sci Total Environ* 44, 51-63.
- Cherian, M. G., Hursh, J. B., Clarkson, T. W. and Allen, J.: 1978, *Arch Environ Health* 3, 109-114.
- Frykholm, K. O.: 1957, *Acta Odont Scand*, Suppl No 22.
- Jokstad, A., Thomassen, Y., Bye, E., Clench-Aas, J. and Aaseth, J.: 1992, *Pharmacol Toxicol* 70, 308-313.
- Langworth, S., Ellinder, C., Göthe, C. and Vesterberg, O.: 1991, *Int Arch Occup Environ Health* 63, 161-167.
- Olstad, M. L., Holland, R. L., Wandel, N. and Hensten-Pettersen, A.: 1987, *J Dent Res* 66, 1179-1182.
- Patterson, J. E., Weissberg, B. G. and Dennison, P. J.: 1985, *Bull Environ Contam Toxicol* 34, 459-468.
- Plewa, J.: 1992, *Int J Risk & Safety in Medicine* 3, 1-22.
- Skare, I.: 1972, *Analyst* 97, 148-155.
- Skare, I. and Engqvist, A.: 1994, *Arch Environ Health* 49, No 5, x-y
- Skare, I., Bergström, T., Engqvist, A. and Weiner, J. A.: 1990, *Scand J Work Environ Health* 16, 340-347.
- Snapp, K. R., Boyer, D. B., Peterson, L. C. and Svare, C. W.: 1989, *J Dent Res* 68, 780-785.
- Stock, A. and Cucuel, F.: 1934, *Z Angew Chem* 47, 641-647.
- Svare, C. W., Peterson, L. C., Reinhardt, J. W. et al: 1981, *J Dent Res* 60, 1668-1671.
- WHO: 1991, World Health Organization task group on "Environmental health criteria for inorganic mercury", Geneva. (Environmental Health Criteria Series No. 118).
- WHO: 1972, World Health Organization/ FAO expert committee on "Evaluation of Certain Food Additives and Contaminants", Geneva. (WHO Technical report series No. 505).
- Vimy, M. J. and Lorscheider, F. L.: 1985, *J Dent Res* 64, 1069-1071.
- Vimy, M. J. and Lorscheider, F. L.: 1985b, *J Dent Res* 64, 1072-1075.
- Åkesson, I., Schütz, A., Anewell, R., Skerfving, S. and Glantz, P. O.: 1991, *Arch Environ Health* 46, 102-109.

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AMSA MERCURY WORKGROUP

AMSA's Mercury workgroup was formed in 1998 to address issues related to the discharge of mercury into the environment and the impact of anticipated mercury effluent limits and sensitive analytical methods to publicly-owned treatment works (POTWs). The workgroup has performed significant work to ascertain the low levels of mercury in POTW discharges and the sources of mercury into the POTW collection system. The workgroup is also working with U.S. EPA to improve the range of analytical methods used to detect mercury in wastewater, and is seeking appropriate modifications to mercury criteria using best available science.

The workgroup plans further study to evaluate the effectiveness of source control/pollution prevention programs to achieve anticipated regulatory compliance levels and plans to explore potential national compliance strategies for mercury so that every individual POTW does not have to come up with an individual compliance solution. Agencies represented on AMSA's Mercury Workgroup include:

Sanitation Districts of Los Angeles County, CA
Orange County Sanitation District, CA
Sacramento Regional County Sanitation District, CA
City of San Diego Metropolitan Wastewater
Department, CA
City of Palo Alto Regional Water Quality Control
Plant, CA
San Francisco Public Utilities Commission, CA
City of Los Angeles Department of Public Works, CA
City of San Jose Environmental Services
Department, CA
Central Contra Costa Sanitary District, CA
Metropolitan Water Reclamation District of Greater
Chicago, IL

Massachusetts Water Resources Authority, MA
Wayne County Department of Environment, MI
Western Lake Superior Sanitation District, MN
Metropolitan Council of Environmental Services, MN
Onondaga County Department of Drainage and
Sanitation, NY
City of Lima Utilities Department, OH
Metropolitan Sewer District of Greater Cincinnati, OH
City of Akron - Public Utilities Bureau, OH
City of Toledo Department of Public Utilities, OH
Northeast Ohio Regional Sewer District, OH
Hampton Roads Sanitation District, VA
Green Bay Metropolitan Sewer District, WI
Milwaukee Metropolitan Sewerage District, WI