Bull. Environ. Contam. Toxicol. (1985) 34:459–468

Environmental Contamination and Toxicology

Mercury in Human Breath from Dental Amalgams

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Silver-tin amalgam, (approx. 50 weight percent mercury) as used in dental restorations, has proven an extremely successful material but its history in dentistry is punctuated with controversy because of the well known health hazards of mercury (WHO 1976, 1980, Gerstner and Huff 1977, Nriagu, 1979). Recent literature has mostly upheld the widespread belief that silver-tin amalgams become inert and release neglible amounts of mercury to the patient once they have fully hardened a few days after placement (Frykholm 1957, Rupp and Paffenbarger 1971, de Freitas 1981). The major area of interest has been the possible exposure of dentists and assistants to mercury vapour in dental offices which may be contaminated (Proceedings International Conf. on Mercury Hazards in Dental Practice 1981, Rao and Hefferen 1982).

Modern methods of mercury analysis have made trace mercury vapour determinations simple and rapid in comparison with the pioneering and definitive efforts of Prof. A. Stock and coworkers (Stock 1941), who actually recovered all the mercury from a sample as droplet of elemental mercury and measured its diameter with a microscope. It is interesting to note that 1 ng of mercury is readily observable as a 5 micron diameter droplet. In spite of the great interest and the large amount of research in this field very few in vivo measurements of mercury in human breath related to silver-tin amalgam fillings have been reported. Those measurements that have been reported have either not been assessed in relation to exposure limits (Svare et al., 1981) or have been largely overlooked in recent studies (Stock 1939).

We report here the results of our measurements of mercury vapour concentrations in the exhaled breath of 172 persons, a few of which exceed probable safe exposure limits and appear high enough to be a chronic toxicologic hazard for some people with numerous

amalgam fillings.

MATERIALS AND METHODS

Breath samples were taken from a total of 167 adults with silver-tin amalgam fillings plus 3 adults and 2 children without amalgams. In a subsample of 94 persons (age range 16 to 66 years, mean 37 years and standard deviation 11 years) the number of tooth surfaces containing silver-tin amalgam fillings was counted during a dental examination. Each tooth has a possible five surfaces that may be filled; mesial, distal, lingual, buccal and occlusal. Breath samples were taken from this group and 12 others not examined, both before and after brushing the teeth for one minute with a soft brush and a commercial tooth paste (hydrated alumina abrasive).

Breath samples were collected by exhaling from the mouth, at approximately 2 l/min, through a 2.5 mm ID polypropylene mouthpiece attached to a silica-glass tube (130 mm x 4 mm ID) with 0.5 g of gold turnings fixed in the tube centre. Sample volume was measured by a 1 litre breath alcohol sampling bag attached to the silica-glass tube exit. The subject's mouth geometry was not controlled during sampling although this was later found to be a variable responsible for some anomalously low results, by as much as a factor of six in some cases.

Immediately after sample collection the sample tubes were either sealed for later analysis (about 50 samples collected at a dental surgery, where the ambient air contained 0.2 ng/l) or were analysed immediately in the laboratory (ambient air 0.08 ng/l mercury) by direct connection to the nitrogen carrier gas inlet (oxygen and mercury free at a flow rate of 0.25 l/min) of a twin cell version of a photoacoustic mercury detector (Patterson 1982). The mercury collected on the gold was released by flame heating and determined during the following two minutes. An inlet membrane filter and a calcium oxide granule trap prevented contaminants affecting cell performance. After analysis, the ends of the sample tubes were sealed for re-use. This method of sample handling very effectively excluded contamination of samples by mercury from the ambient atmosphere.

The instrument was calibrated using syringe transfer of air in equilibrium with mercury vapour at a known temperature. Calibrations were constant over long time periods with variations of less than 5 percent at 10 ng. A linear working range from 0.1 to 80 ng was set although in one case this was extended down to 0.02 ng

for a subject with no amalgams, where a 10 l sample was taken yielding a concentration of 0.008 ng/l. This subject was therefore acting as a sink for mercury in our laboratory air which contained 0.08 ng/l. Mercury was also determined in some samples by conventional cold vapour atomic absorption spectrometry, in tandem with the photoacoustic instrument, with identical results.

RESULTS AND DISCUSSION

Our results are summarised in Figs 1 to 3 and in Table 1. In fig 1a a histogram of mercury concentration in exhaled breath versus frequency of occurrence is shown for 167 persons with silver-tin amalgams under typical conditions without disturbing the amalgam surfaces. The increase in mercury levels from before brushing in fig 1b to the levels shown in fig 1c after brushing are for a subgroup of 106 (94+12) persons. In this subgroup 2 persons had no amalgams. Fig 1d shows a frequency distribution of the number of tooth surfaces with silver-tin amalgams, for the 94 persons given dental examinations.

For our total sample of 172 persons before disturbing the amalgam surfaces by brushing, mercury concentrations in breath ranged from 0.2 to 28 ng/l, the mean mercury concentration in breath was 3.1 ng/l, the median was 1.3 ng/l and the top tenth percentile was greater than 8.2 ng/l. After tooth brushing mercury concentrations in breath increased to range from 0.1 to 62 ng/l with the mean increased to 8.2 ng/l, the median was 4.1 ng/l and the top tenth percentile was greater than 20 ng/l for the subgroup of 106 persons shown in fig 1c.

The large increase in mercury concentrations in breath caused by brushing the teeth also occurs after any form of even mild abrasion of the tooth surface such as eating a meal or chewing gum. The enhanced values obtained by brushing the teeth decreased only slowly over the following hour to about one third of their peak value. Eating an egg, however, caused an immediate decrease to pre-brushing concentrations, presumably because of tarnishing of the amalgam surfaces, but the tarnish observed during the dental examinations showed only a poor linear correlation ($r = -0.3$) between an arbitary index (0 = shiny, 7 = black) and mercury in breath.

Our results are in very good agreement with the few measurements of mercury in breath that have been published. As early as 1939, mercury concentrations of

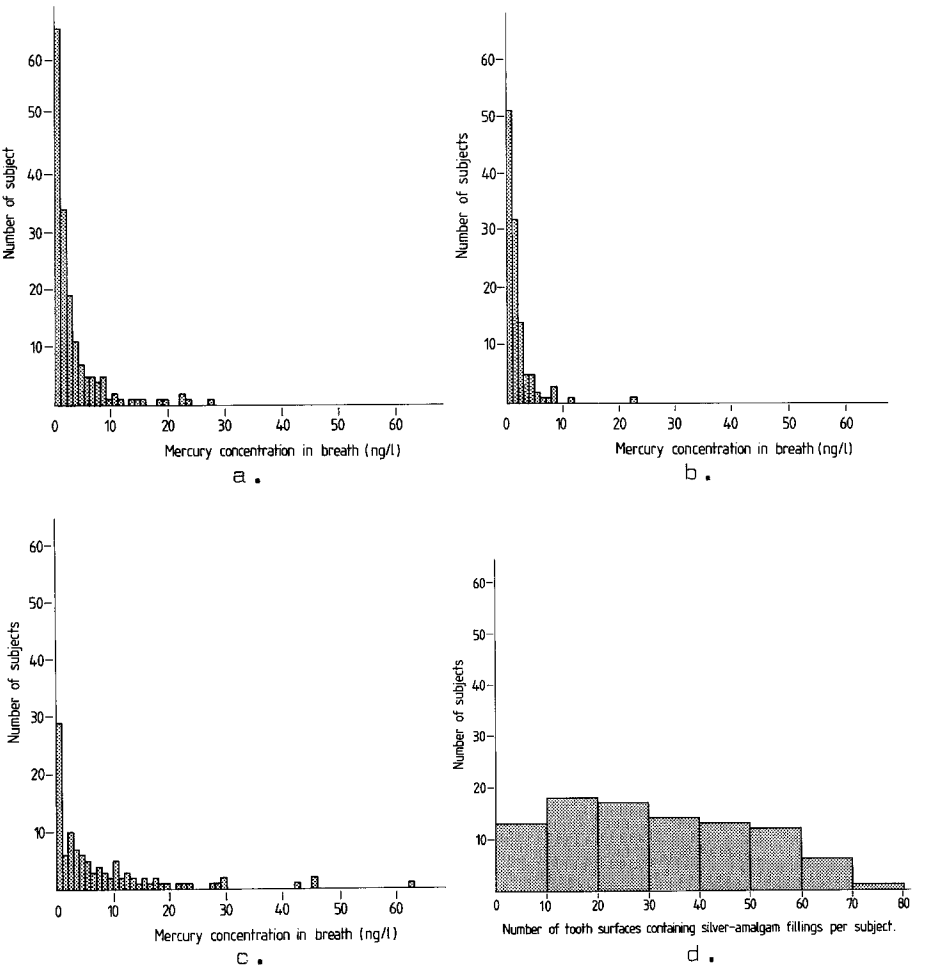

Figure 1. Histograms of mercury concentrations in expired breath:

a. for 167 subjects with silver-tin amalgams (mean 3.19, std deviation 4.78, median 1.30 ng/l)
b. before tooth brushing for 106 subjects (mean 1.92, std deviation 2.93, median 1.00 ng/l)
c. for 106 subjects after tooth brushing for 1 minute with a soft tooth brush and toothpaste (mean 8.17, std deviation 10.9, median 4.1 ng/l)
d. Histogram of the number of tooth surfaces with silver-tin amalgam fillings for each subject (potentially 5 surfaces per tooth) for the 94 subjects given dental examinations.

0.1, 0.2, 1.0, and 2.1 ng/l were reported in the exhaled breath of four persons with silver-tin amalgam fillings by Stock (1939).

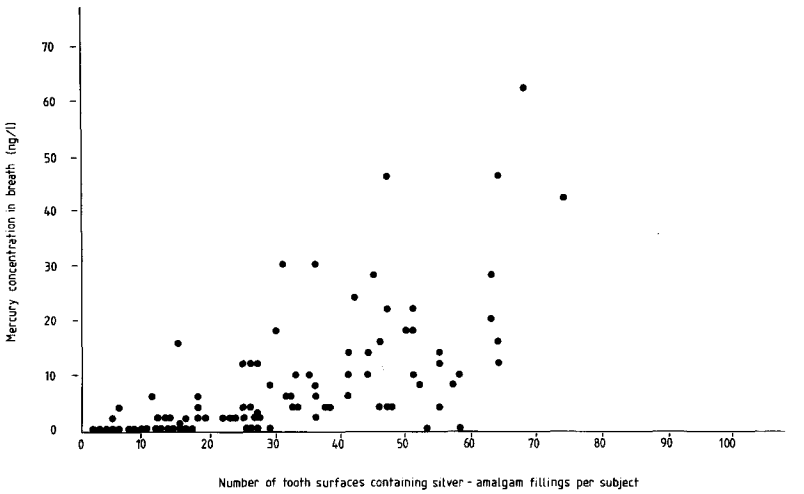

Figure 2. Mercury concentrations in expired breath after tooth brushing versus the number of tooth surfaces containing silver-tin amalgams per subject for 94 subjects. The correlation coefficient is 0.63 assuming a linear model.

Recently Svare et al. (1981) reported a 15 fold increase in mercury in exhaled breath caused by chewing gum. In their research with 40 persons with amalgam fillings, initial concentrations of mercury in exhaled breath ranged from 0.1 to 2.61 ng/l (mean 0.88 ng/l) which increased to a range of 0.08 to 85.5 ng/l (mean 13.74 ng/l) after chewing gum for 10 minutes. For 8 control subjects without amalgams, the mean concentration of mercury in expired breath was 0.26 ng/l before, and 0.13 ng/l after chewing gum.

Observed mercury concentrations in exhaled breath increase with the number of tooth surfaces containing silver-tin amalgams; with $r = 0.41$ before brushing to $r = 0.63$ after brushing (fig 2) if a linear model is assumed. Other dental indices such as an estimate of total amalgam surface area gave similar levels of linear correlation to the simple count of tooth surfaces containing amalgams used here. This correlation coefficient was further strengthened to $r = 0.9$ and a slope just above the upper boundary of the data in fig 2 for a subgroup of 10 persons, selected because they had anomalously low mercury concentrations in their breath after brushing, with their data points lying near the abscissa of fig 2. This higher correlation coefficient was obtained by alternate

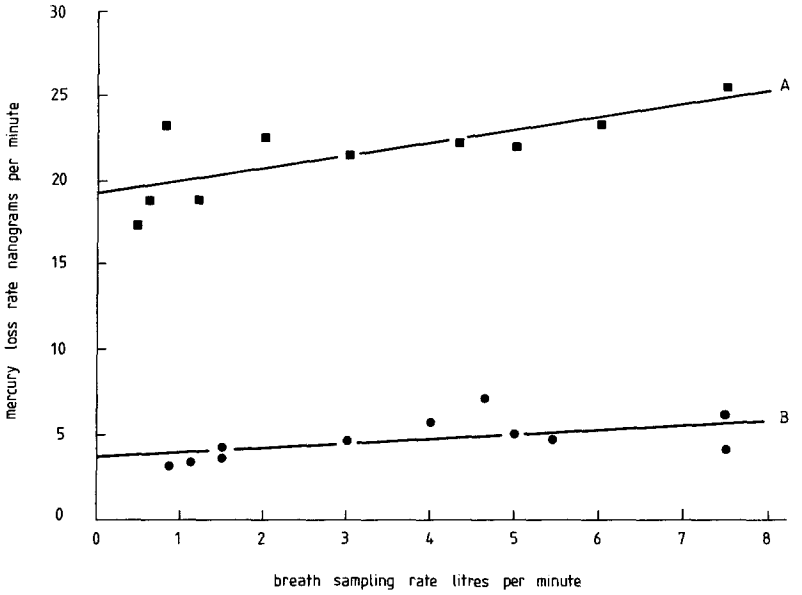

Figure 3. The rate of mercury loss from in situ silver-tin amalgam fillings versus breath sampling rate as determined in 1 liter samples of expired breath from one subject, with 31 tooth surfaces containing silver-tin amalgams, on two consecutive days;
A. three hours after having muesli for breakfast,
B. three hours after having an egg for breakfast.

brushing and sampling until steady maximum values were obtained, usually after 3 or 4 sequences, and by controlling mouth geometry during sampling. This was achieved by the subject holding a 22 mm diameter plastic mouth piece between their teeth while exhaling with dilated cheeks which allowed a more uniform exposure of the teeth to the exhaled breath. The low results obtained for some individuals in fig 2 are caused by a combination of incomplete cleaning of amalgam surfaces and also by incomplete exposure of the amalgams to exhaled breath.

The upper boundary of the data in fig 2 probably represents a potential maximum for each subject (coincidently almost numerically equal to the number of silver-tin amalgam surfaces) if the amalgam surfaces are well cleaned and mouth geometry is controlled during sampling.

TABLE 1. Comparisons of mercury Concentrations from Various Sources

Mercury Conc ng/l		Source	Reference
		BREATH	
0.2-28 (mean 3.1)	167	persons with amalgams before brushing	This work
0.1-62 (mean 8.2)	104 +2	persons with amalgams without after brushing	This work
0.008-0.1 (mean 0.06)	5	persons no amalgams	This work
0.1, 0.2, 1.0, 2.1	4	persons with amalgams	Stock 1939
0.1-2.61 (mean 0.88)	40	persons with amalgams Before chewing gum	Svare et al. 1981
0.08-85.5 (mean 13.7)		After chewing gum for 10 minutes	Svare et al. 1981
	8	persons no amalgams	Svare et al. 1981
0.26 mean		Before chewing gum	Svare et al. 1981
0.13 mean		After chewing gum 10 minutes	Svare et al. 1981
		AIR	
0.08		Ambient, DSIR lab.	This work
0.2		Dental surgery	This work
0.001-0.003		Unpolluted marine air	Slemr et al. 1981
		RECOMMENDED LIMITS	
10-100		Threshold Limit Values of various countries	Gerstner and Huff 1977
25		WHO Recommended Health Based Occupational exposure limit	WHO 1980
7.7		Equivalent time weighted average for continuous exposure at WHO recommended maximum of 25 ng/l	
1		Probable safe limit of continuous exposure for general population	Gerstner and Huff 1977

The rates of mercury release from amalgams are nearly constant over short time intervals (say 15 minutes) for a wide range of breathing rates but vary greatly from day to day depending on the state of the amalgam surface as shown in fig 3 for one person on two consecutive days. To correct our reported values obtained by sampling at 2 l/min to a normal breathing rate of 14 l/min requires division by about six. This factor will overcorrect to some extent where low results were obtained because mouth geometry was not optimum.

Elemental mercury vapour entering the lungs is largely absorbed and passes into the blood stream (Hursh et al 1976). Our measurements are consistent with this. For example breath exhaled from the mouth containing 13 to 40 ng/l mercury at different times yielded only 0.2 to 0.3 ng/l when exhaled through the nose after inhaling through the mouth. Mercury inhaled in breath entering the lungs has not been determined and it is only by assumption that the exhaled breath levels are a measure of intake. Diffusive mixing and local absorption into the blood stream by the oral cavity cannot be excluded for persons who normally inhale through the nose. Daily intakes of mercury at normal breathing rates of 14 l/min would exceed 27000 ng for the upper tenth percentile of our total sample of 172 persons. This is based on our upper tenth pecentile value of 8.2 ng/l, sampled at 2 l/min, normalised to 1.37 ng/l at 14 l/min by dividing by six and a daily breathing volume of 20 cubic metres. Realistic values may be much higher than this estimate because of the frequency of low results caused by poor mouth geometry during sampling (Fig 2).

The central nervous system is the critical organ for the toxic effects of inhaled mercury (WHO 1976). Although elemental mercury in the blood stream is rapidly oxidised, some of it passes the blood-brain barrier before oxidation is complete (WHO 1976). The earliest symptoms associated with chronic low level exposure to mercury vapour are psychological, including loss of memory and mental ability, lack of concentration, tiredness, depression and headaches. Increased exposure leads to insomnia, loss of appetite and weight, shyness, sweating, anxiety, temper etc (Stock 1941, WHO 1976, Gerstner and Huff 1977). The lowest level of chronic mercury vapour exposure where symptoms can be observed is not clearly established. For example Stock (1941) considered that an occupational exposure of 10 to 20 ng/l would usually result in symptoms, while the WHO (1976) associates an occupational exposure level of 50 ng/l with non specific symptoms only in the most sensitive adults. The first specific clinical symptom that is readily

measurable is tremor which may occur at occupational exposure levels of 100 ng/l (WHO 1976, 1980). The WHO (1976) also acknowledges that "although it appears that the occurrence of adverse effects below 50 ng/l has not been unequivocally established, the possible occurrence of micromercurialism at occupational exposure levels below 50 ng/l cannot be ruled out". On this basis, a WHO study group (1980) has recommended a health based occupational exposure limit of 25 ng/l which is equivalent to a continuous exposure limit of 7.7 ng/l (i.e., 154000 ng per day at a ventilation rate of 20 cubic meters per day) for the general population.

Gerstner and Huff (1977) review the establishment of clinical threshold values for occupational exposures to mercury in air that range from 10 to 100 ng/l. They argue persuasively that the clinical threshold value for general populations subjected to continuous exposures should be in the range of 1 to 10 ng/l and that levels below 1 ng/l (20000 ng mercury per day) probably pose no health hazard to chronically exposed general populations.

The highest levels of mercury in breath measured in this study (fig 1a) are comparable with threshold limit values established in some countries, and exceed the probable safe limits for continuous exposure of the general population, as suggested by some workers. We therefore conclude that the levels of elemental mercury in breath derived from silver-tin amalgam fillings represent a significant and undesirable contribution to mans "normal" body burden of mercury. Further development and use of alternatives to amalgam restorations should be encouraged and the potential benefits of antidotes to toxic heavy metals, such as selenium and vitamin E dietary supplements should be clinically evaluated (Frost 1981, Kosta et al 1975, Magos and Webb 1980, Ganther 1980).

ACKNOWLEDGEMENTS. Dr Terry Cutress, Director, Dental Research Unit, New Zealand Medical Research Council, is thanked for providing continued encouragement and the introductions that led to the collaborative aspect of this study. We also thank colleagues and dental patients for their voluntary assistance.

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Received March 5, 1984; accepted May 10, 1984