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Morris B. Jacobs PhD & Leonard J. Goldwater MD

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Absorption and Excretion of Mercury in Man

VIII. Mercury Exposure From House Paint—a Controlled Study on Humans

MORRIS B. JACOBS, PH.D.,† AND LEONARD J. GOLDWATER, MD, NEW YORK

Introduction

THERE HAS been a large increase in the use of mercury-bearing compounds in products and materials with which persons in the ordinary course of life come into contact. Thus mercurials are used very widely as preservatives, fungicides, and bactericides. Such mercurials are incorporated in a host of products ranging from personal use products as toothbrushes and contraceptive jelly to general use products such as air filters and paints. Mercury can also be found in many foods in concentrations up to 10 μ g/100 gm fresh weight.

Foreseeing the widespread and increasing use of latex paints containing antifungal mercurials as preservatives, Goldberg and Shapero, in 1957,¹ studied the toxicological hazards of such mercurial paints. They found that mice living for six months in cages painted with paints containing mercurial fungicides retained no significant amounts of mercury in their kidneys, liver, lung, or spleen. From these results they inferred "that no toxic hazard to humans working in rooms treated with these mercurial paints should result."

Relatively few instances have been reported of poisoning attributable to the in-

halation of mercury vapor in the home. Burke and Quagliana² reported two cases resulting from the inhalation of mercury vapor from an attempt to recover mercury from hearing-aid batteries in a home. These are not really valid examples of "home" poisoning. Mathes et al³ described an instance of mercury poisoning in a family in which a homemade mixture of mercury, turpentine, and aluminum powder was used to paint a gas heater. Citations of this incident have appeared in the literature without qualification so that the inference might be that a commercial paint was used. Another instance of poisoning from mercury vapor in the home was reported by Bucher.⁴ In this case a pound bottle of mercury had been spilled on the rug in the bedroom of a 17-month-old child and even though the mercury had apparently been cleaned up, sufficient mercury remained to yield a harmful concentration of vapor.

In October 1963, Hirschman et al⁵ reported on mercury in house paint as a cause of acrodynia in a 5-year-old boy. In their report, the authors mention the possibility that ingestion accounted for the intake of mercury, but the discussion focuses attention primarily on inhalation. In subsequent correspondence Dr. Hirschman informed us that the child actually had the opportunity of contact (through play) with the mercury-bearing paint almost up until the day he was

Submitted for publication Jan 15, 1965; accepted March 9. From the Division of Occupational Medicine, School of Public Health and Administrative Medicine, Columbia University.

Reprint requests to 600 W 168th St, New York, NY 10032 (Dr. Goldwater).

† Deceased July 12, 1965.

hospitalized. This is a far different picture from that disclosed in the first report.

Although the work of Goldberg and Shapero¹ seemed definitive for animals, it was deemed that the question of the relationship of mercury vapor released from mercury-bearing paints and absorption by human beings was of sufficient interest to warrant study. The prior work that we have performed in the toxicology of mercury has been summarized in a series of papers on various phases of the absorption and excretion of mercury in man.⁶ One aspect particularly applicable to this report is the toxicity of the phenylmercurials which are often used as the fungicides in paint. Thus Goldberg and Shapero worked with paints containing (1) 0.1% w/w phenylmercuric dinaphthylmethane disulfonate, (2) 0.65% w/w phenylmercuric 8-hydroxyquinolate, (3) 0.24% phenylmercuric *p*-tert-octylphenate, and (4) 0.12% phenylmercuric *p*-tert-octylphenate. All of these concentrations were much higher than the concentrations of the phenylmercuric acetate in the paints used in our study.

The objective of the study was to ascertain the degree of mercury exposure and the absorption of mercury that may occur when a paint which contains approximately 0.02% of mercury is used for interior surfaces. Possible effects on the painters who apply the paint and on persons who live in the rooms were studied.

Personnel

The individuals who participated in the study and were exposed to the paint were: AF, painter, man, age 37; RC, painter, man, age 32; HS, occupant of painted room, man, age 61; LT, occupant of painted room, woman, age 47; and NP, chemical analyst, man, age 59.

Three painters, aged 22, 30, and 46 respectively served as controls by painting another room with a nonmercurial paint otherwise similar in composition to that used in the test room.

Prior to the beginning of the study, the subjects were questioned about any possible exposure to mercury. Except for the

previous use of mercury-bearing paint, none of the individuals was aware of any previous exposure to mercury.

Urine samples were collected from all who participated in the study, namely, from the residents of the home, the painters, and an investigator before, during, and after the painting and from two residents for the period of time in which the concentration of mercury decreased to an inappreciable value.

Samples of blood were collected from the painters before they started painting and after they had finished painting. Blood samples were also taken from two residents.

Materials and Procedures

A. General Plan of Operation.—Two rooms in a residence were selected. One of these was a bedroom with approximate dimensions of 12 × 15 × 8 feet, thus having an approximate volume of 1,440 cu feet and the other was a "den" with an approximate volume of 3,000 cu feet. The bedroom was painted with the mercury-bearing paint; the den with the nonmercurial paints. Two painters worked in the bedroom; three in the den. The mercury concentration of the air in the rooms was measured before, during, and after the painting. This was done directly on the premises, with the use of an ultraviolet, photometric, recording mercury vapor meter in the bedroom and an ultraviolet photometric mercury vapor meter in the den. In addition, samples of air were taken by means of pumps, impingers, and trapping solutions and these test samples were subsequently analyzed for mercury in the laboratory.

The mercury vapor meter readings were taken after the painting had been finished until the readings became insignificant.

The ventilation was regulated so as to produce conditions no more favorable for the removal of vapors than would be expected in the normal or usual practice. However, on several occasions the ventilation was restricted to observe any build-up of mercury vapor.

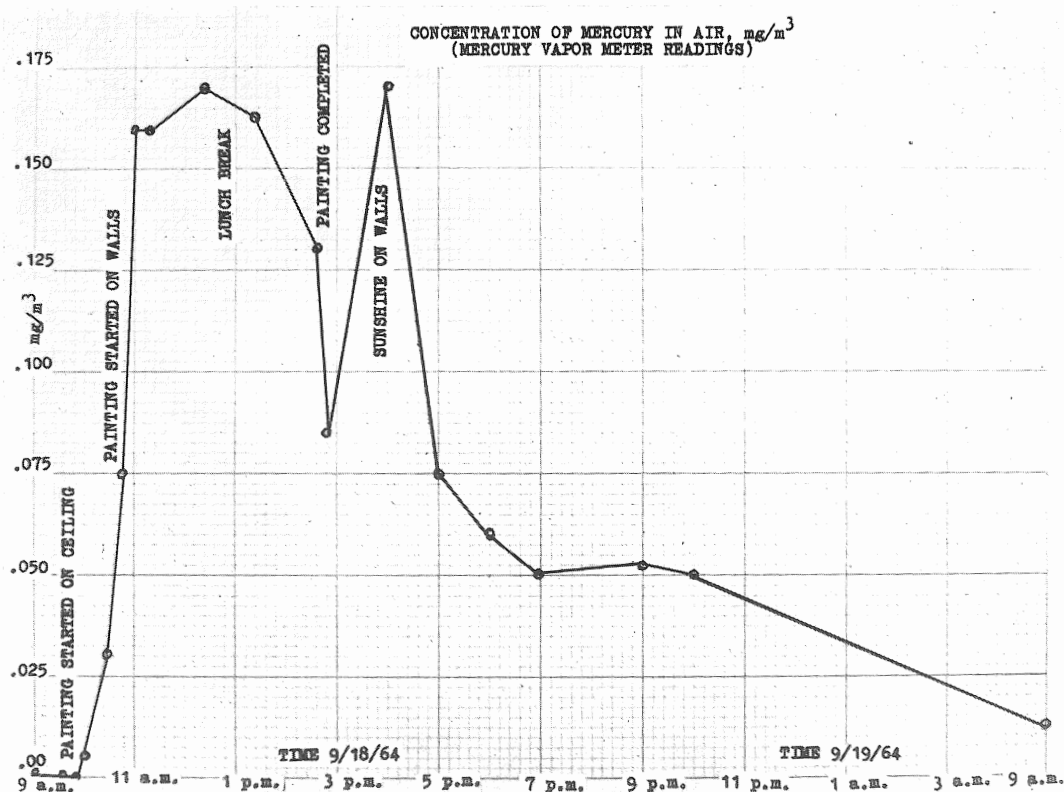
The paints were applied in the customary manner, that is by brush and roller to the walls, ceilings, and woodwork by professional painters.

B. Paints.—1. Wall paint—latex one-coat flat containing 0.02% mercury equivalent to 0.036% phenylmercuric acetate.

2. Ceiling paint—latex one-coat flat containing 0.02% mercury equivalent to 0.036% phenylmercuric acetate.

3. Woodwork enamel—contained no mercury.

4. Wall paint—latex wall paint washable, contained no mercury.



Mercury vapor concentrations found in the bedroom painted with the mercury-bearing paint.

5. Ceiling paint—ceiling flat white, contained no mercury.

6. Woodwork enamel—contained no mercury.

Paints 1, 2, and 3 were used to paint the wall, ceiling, and woodwork respectively of the bedroom, that is, the room to be painted with the mercury-bearing paint. Paints 4, 5, and 6 were used in the control room.

C. Methods of Analysis.—1. Direct Mercury Vapor Measurement: The concentration of mercury vapor was measured directly by means of Beckman Model No. 23 ultraviolet photometric mercury vapor meters. One of these was equipped with a recorder and the mercury vapor concentration was monitored continuously in the room during the painting in which the mercury-bearing paint was employed. The same model mercury vapor meter was used in the room being painted with paint not containing mercury but since this instrument was not equipped with a recorder, a record of visual observations of the meter was made.

2. Total Mercury Concentration: A variation of the Poljaev¹³ and Barnes¹⁴ iodine-iodide method was employed. The principles and general details are given by M. B. Jacobs.¹⁵

3. Urine and Blood Analyses: The concentration of mercury in the urine and blood samples was performed by an ultraviolet photometric method described by Jacobs et al.¹⁶

4. Paint Analysis: The mercury concentration of the wall and ceiling paints was determined by a variation of an ultraviolet photometric method developed by Jacobs for the estimation of mercury in tissues.¹⁷

Results

The results obtained in this investigation are presented in three tables and a Figure. The mercury vapor concentrations found in the bedroom painted with the mercury-bearing paint are given in the Figure. The readings were initially recorded with a recorder but after readings had fallen to a very low value, the use of the recorder was discontinued, and visual meter readings were recorded from time to time.

The readings of the mercury vapor meter in the den are given in Table I. These readings are in all probability attributable to an aerosol effect during the painting period.

The results of the total mercury analyses are tabulated in Table 2. The mercury concentrations found in the urine and blood samples are given in Table 3.

Comment

Examination of the Figure and Table 2 shows that no appreciable amount of mercury was present in the bedroom before the start of painting. Within minutes after the start of painting the mercury vapor meter registered the presence of mercury vapor and rose from 0.00 mg/cu meter at 9:31 AM on Sept 18, 1964 to 0.16 mg/cu meter at 11:00 AM. The total mercury concentration was 0.20 mg/cu meter, that is, it was higher than shown on the meter since mercury as aerosol formed in the painting was trapped in the iodine-iodide absorbing solution. The deposition of very small droplets of paint on the sampling equipment was evidence that aerosol formation occurred. When ventilation was reduced by closing the door of the room there was an evident increase in the mercury vapor concentration. The concentrations of mercury remained higher than 0.1 mg/cu meter until after 4:00 PM even though the painting was finished almost two hours before. When direct sunlight entered the room and shone on the walls, the concentration of mercury vapor in the air increased. The mercury concentration then fell steadily with windows opened slightly and the door open from 4:00 PM Sept 18, 1964 when it stood at 0.1 mg/cu meter to 0.01 mg/cu meter at 9:00 AM the following day. When heat was turned on at 11:00 AM Sept 19, 1964, the concentration of mercury rose slightly to 0.02 mg/cu meter. There was not sufficient present, however, to be detected by the iodine-iodide method (see Table 2).

Examination of Table 1 and 2 shows that no mercury was present in the den before, during, or after the painting. There was a maximum reading on the meter of 0.036 mg/cu meter but this was probably due to an aerosol effect on the meter for four samples taken by the iodine-iodide method showed no mercury.

Examination of Table 3 indicates that there was little if any absorption of mercury by the painters during the period of painting. The absorption of mercury by the other persons who were exposed was signifi-

TABLE 1.—Mercury Vapor Meter Readings* in "Den"

Sept 18, 1964	Readings		Operating Conditions
	Hg Mg/ Cu Meter		
8:55 AM	Set zero & span		Before painting
9:10	0.000		Before painting
10:00	0.000		Start of painting
10:15	0.015		
10:20	0.020		
10:34	0.032		Ceiling almost finished
10:55	0.035		Ceiling finished
11:03	0.030		Considerable of wall painted
11:25	0.020		
11:40	0.022		
12:00 PM	0.036		Still painting walls
12:30	0.019		No painting
1:06	0.000		No painting
1:45	0.000		
2:31	<0.005		Walls finished, woodwork being painted

* These readings are undoubtedly due to paint aerosol interference for the total mercury analyses showed that no mercury was present. Furthermore as soon as the actual painting operations stopped the readings decreased and fell to zero.

cant, as high as 25 $\mu\text{g}/\text{liter}$ in one instance and 30 $\mu\text{g}/\text{liter}$ in the other, but nevertheless of a low order of magnitude. The blood analyses of all persons exposed did not show any significance. The paints said to

TABLE 2.—Air Concentrations of Mercury (Iodine-Iodide Method)*

Sam- ple No.	Location	Time	Hg Mg/Cu Meter	Operating Conditions
1	Bedroom	7:38-8:08 AM	0	Before painting
2	Den	8:30-9:00 AM	0	Before painting
3	Den	10:00-10:33 AM	0	From start of painting through ceiling painting
4	Bedroom	9:45-10:20 AM	0.20	From start of painting to end of ceiling painting
5	Bedroom	10:45-11:15 AM	0.19	From finish of ceiling to finish of wall painting
6	Den	10:55-11:25 AM	0	Ceiling finished, much of wall painting finished
7	Bedroom	1:55-2:25 PM	0.21	Immediately after finish of wall; woodwork being painted
8	Den	2:01-2:31 PM	0	Walls finished; woodwork being painted
9	Bedroom	11:25-11:55 AM	0	Next day after painting, door and windows open

* Pittsburgh Latex paint containing a mercurial preservative used for wall and ceiling in bedroom; Super Kem Tone Latex paint containing no mercurial preservative used in den; the enamels used for the woodwork did not contain mercury.

TABLE 3.—Concentration of Mercury in Blood and Urine

Name	Exposure Period	Date, 1964	Time	Blood Hg μ g/100 MI	Urine Hg μ g/Liter
Exposed					
AF	Before	9/18	9:00 AM	0	4.0
	During		11:00 AM	—	0
	During		1:10 PM	—	4.5
	Finish		2:10 PM	—	0
	After		2:30 PM	0	
RC	After	9/19-20			4.5
	Before	9/18	9:00 AM	3.00	10.5
	During		11:20 AM	—	0
	During		1:10 PM	—	0
	After		2:30 PM	2.40	0
HS	After	9/19-20			27.0
	Before	9/17		—	0
	Before	9/17		0.90	
	After	9/18	2:00 PM	—	0
	After		7:00 PM	—	15.0
	After		12:00 Mid	—	19.5
	After	9/19-20	24 Hr	—	25.5
	After	9/21	24 Hr	—	15.0
	After	9/21		0.75	—
	After	9/21	9 AM-4 PM	—	21.0
	After	9/21-22	7 PM-8 AM	—	4.0
NP	After	9/22	8 AM-4 PM	—	3.0
	After	9/22-23	7 PM-9 AM	—	0
	After	9/23	9 AM-3 PM	—	15.0
	Before	9/17	1:00 PM	—	0
	After	9/18	3:10 PM	—	30.0
	After	9/24		—	40.0
	After	10/1		—	0
LT	Before	9/18	8:30 AM	2.10	3.0
	After	9/19	11:00 PM	—	0
	After	9/20	AM	0.75	0
	After	9/21	8:00 AM	—	0
Unexposed					
AB	Before	9/18	9:00 AM	0	0
	After		2:30 PM	0	0
DE	Before	9/18	9:00 AM	0	0
	During		12:25 PM	—	3.0
	After		2:30 PM	0	3.0
GF	Before	9/18	9:00 AM	3.00	0
	During		1:10 PM	—	4.0
	After		2:30 PM	0.30	0

contain mercury were found to have mercury by chemical analysis and those described as not containing mercury did not have any.

In view of the interest aroused by the publication of the article on mercury in house paint as a cause of acrodynia by Hirschman et al⁵ and the inference that the acrodynia could have resulted from the inhalation of the mercury vapor emitted during and subsequent to the painting, we thought we might test this inference by calculation. Doubt has been cast on this inference by Eckardt¹⁸ and in our opinion this doubt was not dispelled by the reply of Hirschman and his collaborators.¹⁹

In comparing our results of the air of an actual painting job with the panel and jar dynamic experiment used by Hirschman et al, it is interesting to note that they obtained a value of 0.17 mg of mercury per cubic meter in 30 minutes whereas we obtained the same concentration in 90 minutes. Their calculated average concentration of mercury was 0.21 mg/cu meter for a six-hour span (on the basis of their corrected values in *New England J Med*).¹⁹ The average value we found for the total painting period of 5½ hours was 0.20 mg/cu meter. No comparison can be made with their value of 0.76 mg/cu meter under static conditions since our room was painted with the door open and the windows partly open. One must note, however, that there were increases in the room concentration of mercury when the door was closed, when sunlight was on the walls, and when the heat was turned on.

Although a tentative calculation could be made of the apparent amount of mercury vapor breathed by EL for a given period, say two days, the amount excreted could not be calculated since the values shown in the Hirschman et al graph are concentrations corrected on the basis of specific gravity and therefore have no meaning. Furthermore as already noted, Hirschman's subsequent letters indicate that the boy was exposed for a much longer period of time than indicated in his paper.⁵

One can only conclude that EL did not inhale a sufficient amount of mercury vapor to cause the acrodynia and undoubtedly absorbed the mercury either by ingesting some paint or by absorption through the skin from paint on the hands, arms, face and other exposed parts of the body.

Summary and Conclusions

1. When paints containing a mercury-bearing preservative were used, mercury vapor was elaborated. It reached a value of 0.17 mg/cu meter in about 90 minutes. It stayed at this concentration level for about two hours and then fell to 0.01 mg/cu meter in 24 hours. The total mercury con-

centration was of the order of 0.20 mg/cu meter for about 4½ hours.

2. After 24 hours with no exceptional attempts at ventilation the concentration of mercury decreased to an insignificant level.

3. Some mercury was absorbed by persons exposed to the vapors. Urinary concentrations were no greater than those found in unexposed "normal" persons.

4. Painters using mercury-bearing paints showed no evidence of absorption or effects of inhaling the concentrations of mercury found in the workroom air.

5. No evidence was found of mercury exposure or absorption in a degree that would constitute a hazard to the painters or to the occupants of the painted room.

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