On The Potential Hazard Associated With The Introduction of Tetramethyllead As An Antiknock Agent Into Commercial Motor Gasoline, With Respect To The Absorption of Lead By the Handlers of Such Gasoline

- An Interim Report Made After Nine Months of Distribution -

by

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Introductory

Certain questions arose initially concerning the potential hazard of lead absorption in connection with the distribution and use of gasoline containing the antiknock agent, tetramethyllead. The primary factor which might give rise to hazard through the substitution of this compound for tetraethyllead in equivalent concentration in gasoline (equivalent in terms of the proportional representation of lead in the molecule, whereby, at the maximum, 2.06 ml. of TML are equivalent to 3.00 ml. of TEL), was that of the appreciably greater volatility of tetramethyllead. The practical question revolved around the extent to which this higher volatility might operate with significant effect to contaminate the air with tetramethyllead, or whether this factor would be offset sufficiently by the high dilution of the tetramethyllead in the gasoline, and, additionally and more pointedly, by the movement of currents of air in diluting any such volatilized lead to insignificant levels of concentration in the air breathed by those who would handle the gasoline under the ordinary, as well as the possibly extraordinary (from time to time), conditions of the commercial handling of the gasoline.

This question, insofar as a comparatively short period of time and a specific set of conditions (regarded as uncommonly severe) of use were concerned, had been answered in the negative (See Report of The Kettering Laboratory of November 10, 1959). It was believed, however, that the matter was of sufficient importance to justify further investigation of the conditions associated with the gamut of commercial operations over a period of time sufficient to reveal any even slight increase in the occupational exposure to lead of the handlers of such gasoline. (It has been shown that persons subjected to varying degrees of exposure to lead, in the course of occupations that follow approximately the normal schedule of 40 hours per week of employment in the U.S.A., reach a practical equilibrium with their environment, with respect to their absorption and excretion of lead, as demonstrated by the concentration of lead in the blood, and by the rate of the urinary excretion of lead, in from five to eight months, after which no further increase will occur so long as the conditions of exposure remain essentially constant.) Accordingly, a program of investigation, to be carried out over the period of one year, was designed along two general lines - one concerned with the periodic measurement of the lead in the air in the areas in which men were carrying on various relevant occupations, and the other involving the determination of the approximate rate of their urinary excretion of lead from time to time. (Determinations of the lead content of the blood of the workmen were to be made at the beginning and at the end of the investigation, so as not to ignore any evidence of the absorption of lead in any form. However, the absorption of tetramethyl lead cannot be detected by determinations of lead in the blood, but, rather, is revealed by the measurement of the lead in the urine. The latter, therefore, was to be followed at intervals of three months throughout the year, so as to assess in quantitative terms any degree of occupational exposure to tetramethyllead that might be incurred by individuals and groups.)

Investigative Methods

Inasmuch as a final report of this investigation is to be made in full detail on its completion, it may suffice for present purposes to deal briefly with the experimental design and the methods of procedure. A preliminary survey distributed, resulted in the selection of the sites at which air analyses would be carried out systematically, and in the selection of various groups of employees, whose exposure to the potential hazard was regarded as representative of the various types of occupational exposure in their most severe form. Certain methods of the environmental survey will be described briefly in the following Part 1 of this report, while the general procedure with reference to the employees is outlined in the following paragraphs.

Men engaged at the various occupational sites were selected for periodic investigation, according to the numbers of such persons available, or in such numbers as would be representative of their occupation. When the total number of available persons was ample, every effort was made to select those employees whose service could be expected to continue uninterruptedly (steadiness and reliability), as well as those whose work appeared to involve the maximum severity and duration of exposure. The Table below indicates the sites, the specific operations involved, and the types and numbers of personnel projected for investigation in the San Francisco area:

Sites	Types of work	Numbers of Workmen
Refinery		
1. Blending plant	Regular mixing of TML with gasoline	5
2. Tank Truck Loading Rack	Loading petroleum products, including gasoline containing TML. Approximately 60 drivers have irregular, intermittent exposure, but only the loaders, 12 in number, are exposed regularly.	12
3. Wharf	Loading barges, inspecting, sampling and testing contents	4
4. Barrel Filling Area	Workmen fill drums with gasoline containing TML. 2 are the regular employees and 3 others are available for relief	5
5. Chemical	Testing and distillation of gasoline.	

Sites	Types of work	Numbers of Workmen
Service Stations in S.F. 10 very busy and widely distributed in area	Regular dispensing of gasoline to consumers. Some turnover of persons expected. Substitutions then to be made	Approximately 30

Comparable (somewhat larger) numbers of persons employed in the foregoing categories, with the exception of barrel filters (no such operation was carried out at the refinery in the Los Angeles area, was selected. The actual numbers of persons in the various groups will be shown later in the tabulation of the analytical data.

Each individual in these groups was examined by a physician associated with the medical department of the oil company concerned. The details of these examinations will be reported later after the final examination has been carried out. The examination consisted of a medical and occupational history, a complete physical survey, a hematologic examination, and certain other laboratory procedures according to routine or as required in specific instances. At the initial examination, duplicate samples of blood for analysis for lead were obtained, together with a sample of urine, collected in containers supplied by The Kettering Laboratory and in accordance with the instructions of the Laboratory. All of such samples were forwarded to The Kettering Laboratory for analysis.

Additional samples of urine were collected at intervals approximating three months and forwarded to The Kettering Laboratory. A terminal series of samples of urine is to be collected in May and June of 1961, at which time a second and final series of samples of blood will be obtained. The findings obtained by the analysis of the urine of the employees, together with certain general comments, will be presented in Part 2 of this report. All of the results will be dealt with in a final summary. While the initial analytical results on the blood of these persons is of considerable interest in another relationship, they have little usefulness at the present time for the purposes of this report. Nor will they acquire significance until the terminal series of samples has been obtained and analysed. Therefore, the available data relative to the blood are not presented at this time.

Part 1. Lead In The Atmosphere In Association with Certain Industrial and Commercial Operations.

The operations of interest, located in the San Francisco and Los Angeles areas, included the blending of basoline with antiknock compound containing tetramethyllead, the filling of tank trucks with such gasoline at refinery loading racks, the filling of barrels with gasoline, the testing of the gasoline by various methods (distillations etc.) in refinery laboratories, and the dispensing of the gasoline at filling stations.

The environmental conditions associated with these operations, with respect to the concentration of lead in the atmosphere to which workmen might be exposed, are being investigated systematically in a program of sampling and analysis. Two petroleum refineries, one in each area, provided the sites of two sets of operations for investigation. In addition, twenty-one gasoline service stations were included, ten in the San Francisco area, and eleven in the Los Angeles area. These stations were selected on the basis of the large quantities of gasoline dispensed and their geographic distribution within the two cities.

In order to obtain some clue to the impact, if any, of the use of tetramethyllead upon the level of lead concentration in the atmosphere in metropolitan areas, as compared with the results of observations made prior to its use, systematic determinations are being made of the concentrations of particulate lead in the air at one station in each city. The sampling sites were selected to represent general conditions near the centers of the downtown commercial areas of each city.

Procedure

The same sampling equipment and procedures for collecting the samples are being used in the environs of the two cities, all tests being conducted by technical personnel of the two major suppliers of tetramethyllead, in accordance with procedures agreed upon in advance. The representatives of but one of the suppliers are responsible for carrying out the sampling program in a single area. There is an inevitable degree of variability in the performance of the two groups, and also in the characteristics of the sites of sampling in the two areas. Such variables will be commented on to the extent that they seem worthy of note.

Organic lead is being determined by means of specially designed equipment furnished by one of the suppliers. In initial tests, four cubic feet of air were drawn through the collecting equipment at the rate of 0.5 cubic foot per minute. However, as a result of an investigation of the chemical properties of vaporized tetramethyllead, it was found advisable to reduce the rate of sampling to 0.25 cubic foot per minute, and to use two iodine scrubbers in series. The lead retained in each scrubber is being determined directly in the field, and the sum of the two values is being recorded as the quantity of organic lead per cubic foot of air.

The rate of sampling at the service stations is the same as that used in the other test areas, except that the total volume of air sampled is 8 cubic feet. Before air enters the iodine scrubbers, it passes through a millipore filter, which removes the particulate lead from the air. The concentration of organic lead is determined directly in the field, while the millipore filters are shipped to The Kettering Laboratory in Cincinnati for the analysis of their content of inorganic lead.

The levels of concentration of particulate lead compounds in the atmosphere in commercial centers of Los Angeles and San Francisco are being determined by the analysis of material collected on glass-fiber filter webs used in Staplex high volume samplers. The material from about 2000 cubic meters of air is being removed from the air by each filter during each 24 hours of sampling. The filter webs are being shipped to Cincinnati for analysis.

The following frequencies of sampling are being employed:

1. Blending plants and loading racks are visited every other week.
2. Other samples in refineries (from laboratories and barrel filling operations) are being collected as opportunities present themselves in connection with the active performance of the operations. These samples are being analyzed only for their content of organic lead.
3. Each filling station is being visited for sampling two times per month. These samples are analyzed for their content of both inorganic and organic lead compounds.
4. Samples of particulate material from the general atmosphere are being collected every other day and are being analyzed for their content of inorganic lead. (Adequate evidence has demonstrated the futility of the search for organic lead in the atmosphere of such areas.)

<u>Results</u>.

The results of the environmental survey, as reported herein, derive from samples collected up to February 2, 1961 in San Francisco, and up to February 9, 1961 in Los Angeles.

Tables 1 and 2 give the mean concentrations and the frequencies of occurrence of certain ranges of concentration of lead in the air at the various locations in the refineries of the two areas. In Tables 3, 4 and 5 are summarized the results of the analysis of samples collected at the filling stations in San Francisco, Los Angeles and Bakersfield. The levels of

Table 1

The Frequencies of Occurrence of Various Levels of Concentration of Organic Lead in the Air, within the Ranges Indicated, at Certain Locations in a Refinery in the San Francisco Area

Ranges mic.g.per cu.ft.	In Blending Area: At Dome of Car	In Blending Area: At Weigh Tank	In Blending Area: Outside	At Loading Racks: On Top of Truck	At Loading Racks: On Ground	Barrel Filling Area	Inspection Laborator on Wharf
0-0.499	2	15	3	15	18	3	3
0.5-0.99	8	1	8	20	4		1
1.0-1.49	5	1	1	7	1		
1.5-1.99	3	1	5	4			
2.0-2.49			3				
2.5-2.99	1	1	2	3			
3.0-3.49	3						
3.5-3.99	1						
4.0-4.99	1						
4.5-4.99	2		1				
5.0 and More	14						
Number	40	19	23	49	23	3	4
Mean	5.44	0.29	1.33	0.64	0.14	0	0.13
Std. Dev.	7.39	0.68	1.47	0.65	0.27		

Table 2

The Frequencies of Occurrence of Various Levels of Concentration of Organic Lead in the Air, within the Ranges Indicated, at Certain Locations in a Refinery in the Los Angeles Area

Ranges	Blending Area			Loading Rack	
mic.g. per cu.ft.	At Dome of Car	At Weigh Tank	Ground Outside	Top of Truck	Ground
0-0.499	7	8	14	2	2
0.5-0.99	3	6	2	5	5
1.0-1.49	1	2	1	4	3
1.5-1.99	2		1		2
2.0-2.49	1			2	
2.5-2.99		1			
3.0-3.49					
3.5-3.99					
4.0-4.49	3	1			1
4.5-4.99					
5.0*	1*				
Number	18	18	18	13	13
Mean	1.53	0.74	0.22	0.85	1.00
Std. Dev.	2.00	1.27	0.41	0.58	0.99

*7.00

Table 3

Levels of Concentration of Lead in the Air at Filling Stations in San Francisco

Ranges mic.g. per cu.ft.	Total Lead	Organic Lead	Inorganic Lead
0.000-0.099	65	119	89
0.100-0.199	32		55
0.200-0.299	17	20	10
0.30	18		9
0.40	4		3
0.50	8	19	1
0.60	11	2	
0.70	3	1	1
0.80	3		
0.90			
1.00+	7	7	
Number	168	168	168
Mean	0.260	0.150	0.110
Std. Dev.	0.305	0.109	0.109

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Table 4

Levels of Concentration of Lead in the Air at Filling Stations in Los Angeles

Ranges mic.g. per cu.ft.	Total Lead	Organic Lead	Inorganic Lead
0-0.099	110	167	134
0.10	43	18	43
0.2	20	9	9
0.3	7		2
0.4	11	1	6
0.5	2	1	1
0.6	2		2
0.7	1		1
0.8			
0.9			
1.0+	2	2	
Number	198	198	198
Mean	0.132	0.045	0.085
Std. Dev.	0.18	0.14	0.12

Table 5

Levels of Concentration of Lead in the Air at a Filling Station in Bakersfield

Ranges mic.g. per cu.ft.	Total Lead	Organic Lead	Inorganic Lead
0-0.099	4	16	5
0.1	6	1	8
0.2	1	1	1
0.3	3		1
0.4	2		2
0.5	1		1
0.6	1		1
0.7			
0.8			
0.9			
1.0+	1	1	
Number	19	19	19
Mean	0.28	0.078	0.198
Std. Dev.	0.27	0.23	0.18

concentration of lead in the atmosphere in commercial centers in the two cities are compared in Table 6.

General Comment.

In all of the tables except Table 6, the concentration of lead is expressed in micrograms of lead per cubic foot of air, because of the design and calibration of the equipment employed. In converting them to the more familiar expression of micrograms per cubic meter of air, it is necessary to multiply these values by 35.3, the number of cubic feet in a cubic meter of air.

As may be seen from the data in Tables 1 and 2, the highest levels of concentration of organic lead were found in the air when the domes of the tank cars used in transporting the antiknock compound were first opened for connection to the lines leading to the weigh-tank , into which the compound is transferred at a refinery. A much higher average level of concentration at this location was reached in San Francisco than in the same location in the Los Angeles area. The concentrations fluctuated considerably, depending upon the condition of the valve connections, and on the promptness with which the air sampling was started. The higher average concentrations of lead in the air at the dome of tank cars in the blending area of the San Francisco refinery are regarded as being due to the fact that the procedure of sampling began as the dome was being opened and the connections to the weigh-tank were being made. In Los Angeles, the sampling was initiated, usually, only after connections to the weigh-tank had been made. The two sets of concentrations are representative, therefore, of two different periods of potential exposure at the dome of the car. The highest concentration, ~~of~~ 35 micrograms per cubic foot, equivalent to 1235.5 micrograms per cubic meter of air, was obtained in association with a leak in the dome of the tank car containing the antiknock compound. The actual exposure of the operator under these circumstances is

Table 6

The Frequencies of Occurrence of Various Levels of Concentration of Lead in Air, within the Ranges Indicated, in Los Angeles and San Francisco

Ranges mic.g. per cu.m.	Los Angeles	San Francisco
0-0.99	0	11
1-1.99	6	39
2-2.99	19	16
3-3.99	24	6
4-4.99	16	3
5-5.99	14	
6-6.99	17	
7-7.99	1	
8-8.99	4	1
9-9.99	1	
10-10.99	1	
11-11.99	4	
12-12.99	1	
13-13.99	0	
14-14.99	1	
15 and More	4*	
Number	113	76
Mean	5.3	1.9
Std. Dev.	3.06	1.21

*15.1, 16.8, 17.1 and 21.6

KE 0002888

negligible, since he makes prompt use of his approved canister respirator at the slightest evidence of a leak of the brightly colored fluid into the dome.

Other operations in the process of mixing antiknock compound with gasoline, do not, as a rule, contaminate significantly the air within the building used for this purpose, nor do they contribute significant quantities of organic lead to outside air of the area immediately adjacent to this building (there is a vent line to the outside, rising well above the level of the roof). It will be noted that the air outside of the blending building in the San Francisco area contained more lead than that within the building at the weigh-tank. The reason for this was not ascertained, but it may have been due to the direction of air currents in association with the location of the tank car or the position of the vent, during the unloading operation.

The levels of concentration of lead in the air at the loading racks were generally in good agreement in the two areas. Average concentrations on top of the trucks were 0.64 and 0.85 micrograms per cubic foot, respectively, at the San Francisco and Los Angeles sites. At the Los Angeles operations, the average concentration at ground level was slightly higher than that at the top of the truck. The reason for this is not self-evident, but it is not necessarily remarkable. Trace quantities of lead were found at ground level at the loading rack in the San Francisco area.

The air was sampled during the filling of barrels with gasoline containing tetramethyllead on only three occasions. (The filling of barrels with any one brand of gasoline is not a frequent operation). Organic lead could not be detected in the air during any of these periods. Twenty samples collected in the chemical laboratory were also negative. A trace of organic lead was present in the air of the inspection laboratory on the wharf (locally designated as the "long" wharf) at this refinery.

Small quantities of lead were found in the air at filling stations. In the San Francisco area, the total lead averaged 0.26 micrograms per cubic foot (9.2 microgram/M^3). Of the total lead, 43 per cent, or 0.11 microgram per cubic foot (3.9 microgram/M^3), was inorganic, and 0.15 microgram per cubic foot (5.3 microgram/M^3), or 57 per cent, was inorganic in nature. In the Los Angeles area the total lead in the air at the filling stations averaged 0.132 microgram per cubic foot (4.7 microgram/M^3). The inorganic lead constituted 64 per cent of the total lead present (0.085 microgram/cu.ft), equivalent to 3.0 micrograms per cubic meter of air. The organic lead averaged 36 per cent of the total, being 0.045 microgram per cubic foot, or 1.6 microgram per cubic meter, of air.

At one station in Bakersfield, where 19 samples have been collected to date, the average total concentration of lead was 0.28 microgram per cubic foot of air, equivalent to 7.9 micrograms per cubic meter. Inorganic lead averaged 70 per cent of the total, or 5.5 micrograms per cubic meter.

The higher average concentration of organic lead in the air at filling stations in San Francisco,as compared to those in Los Angeles, is due to a difference in the manner of obtaining the samples. In Los Angeles the air is being sampled near the fueling islands, but not in the immediate vicinity of the gasoline pumps. In San Francisco, the intake of the sampler has been located at approximately the same level as the opening into the fuel tanks of cars, and at a distance of about two feet from the opening. The samples obtained in San Francisco would seem to have contained higher concentrations of organic lead than those in the air to which the attendants are likely to be subjected in the ordinary performance of their duties. It may be that the samples taken in Los Angeles have tended to portray somewhat lower levels of exposure than those which may occur at times. This is, of course a matter of judgment, and, therefore, it is just as well that the two

KE 0002890

sets of data should supplement each other in presenting certain variable possibilities for exposure to vaporized tetramethyllead at service stations.

The average concentration of inorganic lead in the air at the general sampling station in Los Angeles (5.3 microgram/M^3) is approximately three times as great as that in the air of the corresponding station (1.9 microgram/M^3) in San Francisco. The average values obtained in the present investigation may be compared with those of 5.2 microgram/M^3 and 2.4 micrograms/M^3 as found in Los Angeles and San Francisco, respectively, in 1954 by Chambers and associates (Chambers, L.A., Foter, M.J., and Cholak, J. "A Comparison of Particulate L adings in the Atmospheres of Certain American Cities), Proc. 3rd Natnl. Air Poll. Symp., Pasadena, Cal., April 18-20,, 1955; 24-32). Additional information dealing with the impact of the increased use of motor fuels on community air pollution may be gained by comparing the average concentration of 5.3 microgram/M^3, as observed in the present investigation, with that of 6.75 micrograms/M^3, as found in August to November of 1954 in Los Angeles. (Cholak, J., Schafer, L.J., Yeager, D.W. and Kehoe, R.A., Chap. 8, "The Nature of the Suspended Matter, Report #9, An Aerometric Survey of the Los Angeles Basin Aug. - Nov., 1954, N.A. Renzetti, Editor, Air Pollution Foundation 1955).

The foregoing comparisons, while not necessarily or wholly valid, are of more than incidental interest, since the trend suggested by them is contrary to predictions based on calculations from contemplated increases in the numbers of motor vehicle and in fuel consumption, which have been given nationwide publicity. The results of the current investigation, while limited to a single central station in each city, gain something in potential significance in their lower relationship to the earlier results, because of the correspondence of this trend to that clearly established by the data collected in Cincinnati from 1946 through 1959. Several factors were responsible for a decrease in the lead content of the atmosphere in the basin of Cincinnati, including an overall reduction of smoke, the building of expressways, the expansion of populations into the outlying area, and the opening up of the basin of the city, all of these tending to increase the dilution of contaminants. (Cholak, J., Schafer, J.L. and Sterling, T.D., The Lead Content of the Atmosphere. To be published in Journal of the Air Pollution Control Association.)

Part 2. The Excretion of Lead in the Urine of Persons Engaged in Certain Industrial and Commercial Operations.

The data obtained by the analysis of the urine of persons in the various categories of employment have been assembled in a systematic manner for comparison in Tables 7,8,9 and 10 which follow directly.

Table 7

The Urinary Excretion of Lead by Men Engaged in Mixing Antiknock Compound (Containing Tetramethyllead) with Gasoline

(Hypothetical opportunity for exposure to concentrated material)

Lead in Urine in Mg. per Liter

Code Identification	Initial Sample*	Second Sample*	Third Sample*
B.1	0.03	0.015	0.015
B.2	0.045	0.015	0.02
B.3	0.015	0.015	0.015
B.4	0.03**	0.015	0.025
B.5	0.03	0.025	0.015
Averages	0.018	0.017	0.018

* Each sample represents a single voiding (under supervision), the three per man having been obtained at intervals of approximately three months during continued employment in the same work.

** Sample too small in volume to be accepted as representative.

Table 8

The Urinary Excretion of Lead by Men Engaged in Handling Gasoline Containing Tetramethyllead in Various Occupations

Loaders of Tank-trucks

Lead in Urine in Mg. per Liter

Code Identification	Initial Sample	Second Sample	Third Sample
L.1	0.018	0.015	-
L.2	0.03	0.015	-
L.3	0.015	0.11	0.03
L.4	0.02*	0.03	0.02
L.5	0.015	0.015	0.015
L.6	0.035	0.07*	0.03
L.7	0.055	0.055	0.05
L.8	0.032	0.06	0.025
L.9	0.045	-	-
L.10	0.02	0.02	0.015
L.11	0.015	0.02*	0.015
L.12	0.030	0.015	0.035
L.13	0.015	0.015	0.015
Averages	0.026	0.034	0.025

Barrel Fillers

Code Identification	Initial Sample	Second Sample	Third Sample
F.1	0.015	0.02	0.015
F.2	0.02	0.025	0.015
F.3	0.035	0.04	0.035
F.4	0.02	0.04	0.02
F.5	0.025	(0.12** (0.02	0.02
Averages	0.025	0.029	0.021

Personnel of Laboratory

Code Identification	Initial Sample	Second Sample	Third Sample
T.1	0.015	0.02	0.02
S.2	0.04	0.035	0.025
S.3	0.03	0.035	0.02
T.4	0.015	0.02	0.02
Averages	0.025	0.027	0.021

Continued next page

Table 8 (continued)

Gaugers of Gasoline

Code Identification	Initial Sample	Second Sample	Third Sample
G.L.1	0.03*	0.015	0.015
G.2	0.05*	0.015	0.02
G.3	0.015	0.02	0.025
G.4	0.02	0.015	0.015
G.5	0.05*	0.015	0.02
Averages	0.033	0.016	0.019

Wharf Inspectors

Code Identification	Initial Sample	Second Sample	Third Sample
W.1	0.01	0.03	0.02
W.2	0.025	0.02	0.015
W.3	0.03	0.045	0.025
W.4	0.015	0.055*	-
Averages	0.02	0.037	0.02

* Sample too small in volume to be accepted as representative.
** This result was checked by the analysis of a second sample, the result being just below.

Table 9

The Frequencies of Occurrence of Various Ranges of Concentration of Lead In the Urine of Service Station Attendants in the San Francisco Area, at Intervals of Apparently Three Months, In Association with the Dispensing of Gasoline Containing Tetramethyllead

Approximately [illegible]

Lead in Urine in Mg.per Liter	Initial Sample	Second Sample	Third Sample
0.01	6	3	11
0.02	8	10	8
0.03	7	4	2
0.04	3	5	2
0.05	1	2	-
0.06	1	1	1
0.07	1	-	-
0.08	-	1	1
0.15	1	-	-
Total Numbers	28	26	25
Mean	0.036	0.034	0.027
St.Dev	± 0.027	±0.016	±0.017

Table 10

The Frequencies of Occurrence of Various Ranges of Concentration of Lead In the Urine of Service Station Attendants in the Los Angeles Area, at Intervals of ~~Apparently~~ Approximately Three Months, in Association with the Dispensing of Gasoline Containing Tetramethyllead

Lead in Urine Mg. per Liter	Initial Sample	Second Sample	Third Sample
0.01	5	11	7
0.02	8	11	10
0.03	9	4	9
0.04	8	1	2
0.05	2	1	
0.06	2		
0.20	1		
Total Number	35	28	28
Mean	0.040	0.024	0.027
St. Dev.	±0.031	± 0.010	± 0.009

In general, the data as presented in the foregoing tables give a clear answer to the question which led to their collection. With a few exceptions, so few indeed as to be regarded as fortuitous or irrelevant, the concentrations of lead in all of the samples of urine of all of the subjects are so consistently within normal limits as clearly to demonstrate that the groups of persons, regardless of their specific occupations, sustained no appreciable absorption of lead during the period of approximately nine months during which they were subjected to periodic investigation.

When all of the analytical results obtained from the investigation of all of the subjects were assembled and separated only with respect to the three periods of sampling, no valid statistical evidence of any abnormal absorption of lead on the part of the group could be found in any period of the observations. The data distributed themselves statistically in strict accord with comparable types of data obtained from entirely normal persons devoid of occupational or other unusual exposure. There was some variation in the data of the three periods, but the variations showed no consistent trend with the lapse of time, and they were well within the limits of physiological variation, as these have been observed in other data involving samples of urine of small volume. A few individual findings deviated appreciably from the general trend of the results and could be regarded as being outside the statistical universe represented by the data as a whole. For this reason, and also because the combined data were composed of results from inhomogeneous groups of employees, the findings were rearranged for more detailed study, according to the occupations and the geographic distribution of the employees. After exploring every relevant combination and relationship, with a negative outcome, they were reassembled into the tables represented above (Tables 7,8,9 and 10) in which all of their characteristics can be seen readily. Only a few comments are called for.

The few aberrant results, four in all, are in no wise remarkable. Samples collected with the most minute care are contaminated from time to time. These results could be discarded as being irrelevant on the basis of their departure from the statistical universe represented by the other data. However, they have been included in the data of the tables, and have also been examined individually and in their specific relationships. The facts concerning them are as follows:-

In Table 8, under "Loaders of Tank-trucks", a result of 0.11 mg. per liter (which is not entirely beyond the ~~extreme~~ upper normal limit of the concentration of lead in a sample of urine of small volume) is obviously aberrant in the statistical sense and is probably invalid, because of the results obtained by the analysis of the first and third samples of this same person, 0.015 and 0.03 mg. respectively, per liter. (The value of 0.07 mg. per liter which occurs in the same table is not to be taken too seriously, from the aspect of any physiological meaning which it might have otherwise, because of the small volume, by which the analytical error is magnified. It is not, of course, remarkably high, but it stands out by comparison with the other values in this table.)

In the same table, under "Barrel Fillers", is a result of 0.12 per liter. This was sufficiently unusual, especially in this small group of men engaged in an occupation that might (under certain unusual circumstances) give rise to a significant degree of exposure, that a further sample was called for and obtained; on analysis it yielded the value of 0.02 mg. per liter. The prior and subsequent samples may be seen to have been 0.025 and 0.02 mg. respectively, per liter. This relatively high finding can be dismissed from further consideration.

In Table 9 is a single result, obtained in the first round of observations, of 0.15 mg. per liter. This result in a service station attendant could not, by any stretch of the imagination, be regarded as evidence of a dangerous occupational condition, and the aberrant character of this finding could be inferred from its unique relationship to the many others in the same category. Accordingly it was not checked by obtaining a further sample within a short time. The results obtained in the second and third samples from this same subject were 0.025 and 0.045 mg. respectively, per liter.

Essentially the same situation as that above is repeated in Table 10, in which a result of 0.20 mg. per liter, obtained on the first round of sampling, was followed by the finding of 0.015 and 0.02 mg. respectively, per liter, in the second and third samples from the same man.

One additional comment may serve a useful purpose in relation to the urinary findings. The idea has been advanced and has gained credence in certain circles, that samples of urine of small volume, such as those voided at one time, do not convey valid information concerning the general level of the urinary excretion of lead. This is true only of individual samples that are of quite small volume because of the unavailability of suitable quantities of water within the body of the person who supplies the sample, or of samples of unusually large volume voided at a time when the throughput of water is unusually high. (Assuming, of course, that the renal function is essentially normal.) In short, a highly concentrated sample of urine will contain an unusually high concentration of lead, while a highly dilute sample will yield the opposite result. Attempts have been made to correct the analytical result to a standard specific gravity, or a standard concentration of some allegedly invariable or less variable constituent of the urine. Thus far, no such correction factor can be held to be valid, even though it may move the aberrant result somewhat closer to the mean or medium value of a related group of results. The correction is especially questionable in dealing with the extremes of concentration, and it cannot be counted on to do more than artificially to reduce the variability of a series of results. This fictitious and often misleading procedure has not been considered for application to the results presented herein. Rather, we have relied upon a broad experience with a wide variety of results obtained by methods comparable to those employed in this instance, which, without artifice, have supplied the background for the differentiation of "normal" from "abnormal" results. Finally it should be said that the comparison of a series of samples of urine of small volume, with samples of large volume from the same individuals, shows the former to be somewhat more widely variable at both ends of the spectrum, i.e., the small samples make a wider universe, but there is little difference, otherwise, in the distribution and the statistical characteristics of the two sets of results. For the purposes of this investigation, the lower incidence of contamination of small samples collected under supervision and instruction, greatly outweighs the advantage which might otherwise be gained from samples of large volume, while the vagaries of variability in the case of individual samples are blotted out by resort to multiple samples from the same individual and by numbers of individual samples obtained from reasonably homogeneous groups of persons.

Summary

The principal facts obtained in this investigation, as carried out thus far, are summarized under the two main headings indicative of the dual character of the investigative approach.

1. Lead in the air at the occupational sites.

(a) The highest levels of concentration of tetramethyllead are being encountered in the areas in which the antiknock compound is mixed with gasoline, at the domes of tank cars, in which the compound is transported to the refinery, when these domes are first opened. Occasional concentrations up to 35 micrograms per cubic foot were found. This situation is fleeting, and it involves no necessary or likely exposure of the operators at these sites. The average levels of concentration at these points in the blending areas in the vicinity of Los Angeles and San Francisco, respectively, were 5.44 and 1.53 micrograms of lead per cubic foot of air. The lead in the air at other locations in the blending areas averaged less than 2 micrograms per cubic foot.

(b) The levels of lead concentration in the atmosphere of the filling stations averaged 0.132 microgram per cubic foot of air in Los Angeles, and 0.26 microgram per cubic foot in San Francisco. The higher level of concentration in San Francisco derives from the relative nearness of the sampling apparatus to the tanks of the cars during their filling. In consonance with this factor, approximately 55 per cent of the total lead in the air at filling stations in San Francisco was organic lead; in Los Angeles, the organic lead approximated but 35 per cent of the total lead in the air at the filling stations.

(c) Samples of particulate matter collected in central commercial sections of the two cities contained average quantities of lead amounting to 5.3 micrograms per cubic meter of air in Los Angeles, and 1.9 micrograms per cubic meter in San Francisco. These levels are somewhat lower than those reported in investigations carried out in both cities during 1954.

2. The significance of the occupational exposure of the personnel.

(a) Groups of individuals engaged in the several occupations involved in the preparation and distribution of gasoline containing tetramethyllead as an antiknock compound, failed to give any evidence of their absorption of lead in the course of the regular performance of their duties over a period approximating nine months. (The response to occupational exposure to lead, in even low and hygienically insignificant levels of severity, occurs promptly in the form of an increase in the concentration and output of lead in the urine. The failure of such a response is indicative of a negligible degree of exposure.)

(b) The numbers of persons engaged in certain of the occupational groups was too small to provide data for statistical treatment, so that comparisons must be made of the individual findings. In no instance was there a progressive increase (or any increase beyond the level of strictly normal variability) in the concentration of lead in the urine of an individual member of these small groups.

(c) The statistical comparison of the levels of lead concentration in the urine of the two groups of service station attendants (in San Francisco and Los Angeles) brings out certain inconsistent differences. These differences, while of statistically significant magnitude in some instances, cannot be regarded as of physiological significance in relation to the hygienic question under investigation, because of their erratic occurrence in time. Moreover, all of the differences noted are within the range of normal or incidental variability and, therefore, must be accepted as being within the range of the deviations associated with the general methods of investigation.

(e) The statistical comparison of the data obtained at the three stages of the investigation from each of the two groups of filling station attendants also reveals certain differences within the individual groups. These differences have the same characteristics as those referred to above, and, therefore, are of no significance in relation to the primary problem of this investigation.

Conclusion

The results of the surveillance of the industrial and commercial activities associated with the handling of an antiknock compound containing tetramethyl lead in a petroleum refinery and with the distribution of gasoline containing this compound of lead, as carried out over a period approximating nine months, have been summarized above. These results coincide with those of an earlier and less prolonged investigation of this problem, in demonstrating that the exposure to lead, specifically to tetramethyllead, under the conditions which prevail in association with these activities, is negligible. The quantities of lead which escape into the atmosphere during the performance of the relevant operations are small, and the contamination of the atmosphere therewith is intermittent and is regarded as of insignificant proportions in terms of occupational hazard. The insignificance of the occupational exposure to lead is fully confirmed by the lack of any evidence of the absorption of lead by the persons who have been engaged in the performance of the various operations. No individual in any of the categories of employment represented in this survey has manifested any physiological response to the environmental conditions of his work in the form of a demonstrable increase in the rate or magnitude of the excretion of lead in his urine. Inasmuch as such a response can be counted on to appear in the event of the occurrence of a significant degree of occupational exposure to lead in any form, the non-occurrence of such exposure in these relationships is clearly evident.

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