

March 3, 1950

Mr. P. L. Magill,
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Stanford, California.

Dear Mr. Magill:

Mr. L. B. Roberts has told me of the questions raised by you at a recent meeting, with the suggestion that you would be interested in having certain further items of information. I am pleased to give you some additional information, and if I can supply you with more later, I should be pleased to do so.

Your question No. 1, as given me by Mr. Roberts, concerns the disposition of lead in the exhaust gas of automobiles. The answer to this question is available only in general terms, and is not available in the precise terms that relate to the newest kind of automobiles. Investigations were made a number of years ago, by equipping automobiles with Cottrel precipitators of high efficiency, and removing from the exhaust stream all but traces of the lead compound. Following the use of this equipment over a certain mileage, engines were torn down and as much as possible of the accumulated lead compounds gathered and analyzed. The results of these determinations demonstrated that somewhat variable quantities of lead compounds remained in the exhaust system, in which they condensed out and accumulated on the sides of the tube. The proportion varied with the newness of the car and the cleanliness of the exhaust system, tending somewhat to reach a sort of equilibrium at somewhere in the neighborhood of 50% of the total lead burned in the fuel. It would be very difficult to say what is the overall picture in this regard, since the quantities deposited in the exhaust system may be as little as 25 or 30%, and as great as 75% of the lead which is in the gasoline. In any case, that which accumulates in the exhaust system is a source of little or no pollution of the atmosphere, since to the extent that it subsequently gets out of the exhaust system it is probably jarred loose and falls in chunks, so to speak, on the street or roadway. I do not have beside me all the data that are available on this subject in relation to actual road use of automobiles. A considerable number of such data are available, and they can be obtained, but actually if one were to establish the facts in terms of present day operations, these experiments would have to be repeated, since automobile engines and exhaust systems differ in a number of respects from those available when the observations were made.

Your question number 2 concerns the nature of the chemical compounds in the exhaust gas. This depends primarily on the quantity and character of the halogen carrier mixed with tetraethyl lead to make the antiknock compound. In automobile fuels, at present, ethylene dibromide and ethylene dichloride are used, and the lead is converted

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to the corresponding halogen salts, in approximately the proportions in which the two occur in their mixture. In addition small quantities of the oxide, sulphate and, probably, carbonate occur, but all of these individually and collectively occur in negligible concentrations. The halogen carrier is introduced into the fluid in quantities slightly in excess of the theoretical required for the conversion of all of the lead to the halogen salts. The extent of the excess has varied from time to time, but there has always been some excess. Moreover the proportion of the bromide in the fluid, as compared to chloride, has varied from time to time over the years, largely in accordance with the approximate temperature of engine operation, since the purpose of the halogen, is to convert the lead to the compound which will be volatilized almost completely out of the engine under the conditions of normal operation.

Your next question relates to particle size. Few data are available on the actual particle size of the lead compound in the exhaust gas. Such observations as have been made from time to time have demonstrated that the particles are exceedingly small, well under 1 micron, and that they settle out very slowly indeed. It is a well known fact from observations that it is easy to obtain lead bromide in highly finely divided form, to a very uniform extent, by the combustion of leaded gasoline. Observations have been made with reference to the passage of the particles through ordinary filtering media, and it is known that only the very best and most efficient filters will stop the passage of these particles. Consequently, although I do not have available adequate data giving the distribution of particle size, it is quite clear from the observations that have been made, both physical and physiological, that the lead in the exhaust gas is in such finely divided form as to behave substantially like a gas itself over moderate periods of time, and indeed in respiratory experiments carried out by Yant and his associates of the Bureau of Mines many years ago in their investigation of this problem, it was established that a large proportion of the lead inhaled in air contaminated in this fashion, is also exhaled. Incidentally, copies of the Bureau of Mines' report of their observations on the study of this entire problem are very difficult to obtain, being, I believe, substantially out of print. I have a few copies which I have held onto, and so far as I know none other are available. I should be willing to loan a copy to anyone who has reason to be interested in these experiments, but so far as I know there are none available for distribution, and I cannot provide any copies other than on the basis of a loan.

Further question has been raised concerning the fate of the excess halogen in the antiknock compound. So far as I know, no observations have been made that would clearly define the precise fate of the halogens. The presumption is that conversion to the halogen acid occurs, but what proportion this is of the total excess, is not known, so far as I am able to discover. Observations have been made that indicate that hydrobromic acid, for example, is present in minute concentration. Moreover, I have heard some individuals claim that

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they could identify the hydrobromic acid. Perhaps this is true, but I have never been able to do it. In any case, the concentration of hydrobromic acid is quite low.

The next question relates to the source of other lead compounds which occur in the atmosphere. This is an interesting question, and the answer to it is far from being complete in all respects. There are, however, a number of significant sources. One source that is important, and is probably the greatest single factor, is that of the combustion of common fuels in urban and in industrial centers. Coal contains appreciable quantities of lead; wood and other fuels of natural origin contain somewhat less, but the result of the combustion of all of these is to release variable quantities of lead into the atmosphere. Soil contains variable but appreciable quantities of lead, being, wherever it occurs, the result of the disintegration of stones in which appreciable quantities of lead are always found. The soil is more or less pulverized on the surface of the earth, and when it is dry it is picked up by air currents and distributed into the atmosphere. Unquestionably there are other sources of lead compounds in the atmosphere, including, of course, the activities that go on in industrial areas. The distribution of lead in the City of Cincinnati is least in the open country around, and greatest in the industrial areas. The lead concentration along highly traveled highways may show some increase above that in other parts of the city, but the quantities here are always, in our experience, less than those found in certain characteristic industrial areas. Actually we have had considerable difficulty in our observations of the occurrence of lead in the atmosphere to find that increment which should be attributed to the contamination of the atmosphere with the exhaust gases of automobiles. That there is such an increment goes without question, but it rather tends to get lost in the other sources. We have thought we might be able to determine its quantitative significance, by measuring the proportion of lead which occurs in the form of lead bromide. Our efforts along this line have been rather scanty up to the present time, but we anticipate following out this lead for our own purposes. In connection with the overall sources of lead in the atmosphere, it will interest you to know that some considerable improvement in the atmosphere of Cincinnati has been obtained in the past year or two, through efforts to control smoke and soot and fly ash. Coincident with this improvement we have noted a significant drop in the lead concentration in the general atmosphere of the community. This is the biggest change that has occurred in the picture over a period of several years, insofar as the lead concentration in the atmosphere generally is concerned, and it is fairly apparent to us that this is a very significant fact. The present overall concentrations of lead in areas other than industrial areas in Cincinnati at present average along .03 mg. of lead per 10 cubic liters of air. Our results of some years ago averaged well above this level, being nearly twice this figure.

In connection with this entire problem, it will interest you to know that we have planned a rather elaborate study of the total lead

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metabolism of human subjects maintained for approximately 7 hours per day on 5 days a week in a carefully measured concentration of a specific lead compound in a known and fairly uniform state of subdivision in the atmosphere. We know of no way in which the problem of industrial lead exposure can be put on a sound basis other than by this type of study. By a systematic study of this type we hope to add that information, on the inhalation side, that we were able to make available on the ingestion aspects of lead absorption.

I have been told that some very considerable interest has arisen in certain circles in Los Angeles over the problem of the lead content of the atmosphere, and presumably that fraction derived from the exhaust gas of automobiles. Recently I heard some comments to the effect that lead bromide might be a lacrimator, and that it might be responsible for some of the eye irritation that has been complained of. Moreover, I have been told that some representations have been made in relation to the potential hazards of lead in the atmosphere, based on certain rather naive calculations of the total amounts of lead available in gasoline consumed in the Los Angeles area, and therefore being distributed into the atmosphere. These questions were all raised a good many years ago, in somewhat the same naive manner. A very considerable amount of work has been done to answer some of the questions, and all of these data could be gathered together for the purpose of demonstrating the approximate significance of the problem. If it became necessary to do this, I should be very glad to render the assistance that I can. I trust, however, that the problem at issue can be considered on a somewhat more realistic basis. We have been working around toward this problem for a number of years, and I believe that we are now so situated that the questions at issue can be answered in a wholly satisfactory manner. While it is a costly undertaking, it is not too difficult to ascertain the facts concerning the actual degree of the contamination of the atmosphere of representative cities in this country and elsewhere, and I feel certain that it will be possible to establish where the limits of safety lie in this regard. We shall then be able to establish the extent of the safety factor that exists at any time in relation to the existing conditions. As you may be aware, we have been following the lead excretion of rather sizable numbers of people over a period of some years. I have no hesitation whatever in saying that the safety factor in community lead absorption, as the results of the contamination of the atmosphere with lead, is satisfactorily large. There may be specific exceptions to this rule, in that persons who live in the immediate vicinity of an industrial plant in which lead compounds are used extensively, may find deposits of lead compounds in the household, as well as lead concentrations in the atmosphere that moves about them, to be above wholly satisfactory levels.

Finally, let me say that if this problem should become one of considerable importance in the Los Angeles area, I should consider it a privilege to have an opportunity to come out and to talk with the people who would be concerned in the problem and in the means of

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attack upon it.

I trust that what I have said above will answer some of your questions, and it seems likely that others may arise. In any case, if you have any further questions, I shall do my best to answer them.

Cordially yours,

Robert A. Kehoe, M. D.

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P. S. I am sending a few reprints under separate cover which may interest you in connection with this problem at issue. There is not much in them that will actually answer your questions, but they will at least demonstrate something of the manner in which we have approached the problem of lead absorption and excretion previously, and will give you a clue to the methods which we expect to use in the study of inhalation as well as further study of lead ingestion. Any observations made on experimental subjects will have to include all of the sources of lead absorption that are now known to exist, and the subjects will have to be followed carefully with respect to the intake of lead in their ordinary diet, during the period of experimentation.

Reprints under separate cover.

Exposure to Lead
The Concentrations of Certain Trace Metals in Drinking Water
Experimental Studies on the Ingestion of Lead Compounds
Lead Poisoning Symposium

P. . S. Some inquiry on this same subject has come to me indirectly from a Mr. Paul Grimes of the L. A. County Smog Control Commission. You may or may not have the means of talking to Mr. Grimes, but if you should, the information you might give him might be helpful to him and to me.

R.A.K.