

Mr. Barclay

INTERNATIONAL LEADING RESEARCH ORGANIZATION, INC.

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222 MADISON AVENUE, NEW YORK, N Y 10017
TELEPHONE 875-6080 JAMES GUO 8181
CABLE ADDRESS: NYL222 NEW YORK

June 13, 1966

To: All Members of the Lead Health and Safety Committee

Re: Proposal from Isotopes, Inc.

Having received a favorable response from you regarding the Preliminary Proposal prepared by Isotopes, Inc., a group of us met with them to discuss the formulation of a detailed proposal. Representatives from the Ethyl Corporation and du Pont, as well as from the American Petroleum Institute, responded to my invitation to meet and spent a most interesting day at the Isotopes, Inc. office. All of those present were most impressed with our discussion.

As you may see from the attached proposal the measurement techniques and facilities that this potential contractor has should provide the basis for developing invaluable information about the results of modern uses of lead on our environment and the animal life on this planet. Since it seems to us that a great many unfounded allegations of lead contamination are being given credence in government, medical and public information circles, we believe it is imperative that our industry obtain the facts. For instance, we believe that much of the lead found in our environment and the human body is of natural origin and should not be ascribed to contamination by man, but so far we have been unable to document our belief.

Please study this proposal carefully and be prepared to vote on provision of funds in the near future. I will be able to send to you a ballot on a specific amount of money shortly, as soon as I have received from the American Petroleum Institute an indication of their financial interest in this study.

Very truly yours,

Don D. Inula

Don D. Fowler

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A PROPOSAL TO STUDY
THE USE OF LEAD ISOTOPE COMPOSITION
AS AN ENVIRONMENTAL TRACER

Technical and Cost Proposal

Prepared for
International Lead Zinc Research Organization
292 Madison Avenue
New York, New York

and

The American Petroleum Institute
1271 Avenue of the Americas
New York, New York



Thomas D. Parks
Vice President - Technical



Robert C. Butler
Vice President - Treasurer

Isotopes, Inc. Proposal No. 66-11
June 8, 1966

Isotopes, Inc.
123 Woodland Avenue
Westwood, New Jersey 07675

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THE USE OF LEAD ISOTOPE COMPOSITION AS AN ENVIRONMENTAL TRACER

I. INTRODUCTION

A recent report by Patterson, concluded from basic material balance and geochemical considerations that man as an industrial animal has severely contaminated his environment and is suffering lead insult one hundred times greater than that present during his biological evolution. The report further concluded that this level of insult, because it was sustained in a period too short for evolutionary adjustment, is a danger to the well-being of a large portion of our population. Such widespread environmental contamination was regarded as particularly serious because it is cumulative and cannot be easily reduced should a critical level be reached.

The above conclusions are in contradiction to a considerable array of scientific opinion that has developed from extensive work in lead toxicology and pharmacology. In general, the most prominent workers have concluded: 1) that primitive man suffered lead insult no less than one-half that presently experienced by industrial man, 2) that there is a well-defined threshold value of lead insult below which man can exist indefinitely without harm, and 3) that the present "normal" body burden (as measured by the lead content of blood) of about one-third the threshold value allows an adequate margin of safety.

Patterson's approach to evaluating the implications of lead contamination in man's environment was largely theoretical, and because there is not sufficient empirical evidence available to either confirm or refute his argument the controversy remains unresolved. The purpose of this proposal is to outline a research program for the development and application of modern mass spectrometric and radioisotope analysis techniques in order to identify the

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sources of lead in man's environment, to compare the present environment with environments of the past and to elucidate the mechanisms by which lead may become available for biological assimilation. By establishing the feasibility of using the isotopic character of lead as an environmental tracer it is hoped that the relative importance of various possible sources of available lead can be established and that a better understanding of the mechanisms of lead transfer will permit more economical alteration of the environment if required.

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II. TECHNICAL DISCUSSION

The Geochemistry of Lead and the Environmental Health of Man

Lead occurs in nature as a dispersed element in various geological environments and, in addition, is commonly found highly concentrated in the form of ore deposits. Lead is well known for its toxicity, but as a dispersed element it is unlikely to be harmful because it is present only in very low concentrations to which man has adapted during the course of his biological evolution. When lead is encountered in high concentration, however, it can be harmful and man must avoid environments characterized by high lead concentrations whether such environments be natural or a product of industrial contamination.

Figure 1. shows some of the routes by which lead may become available to man from the primary sources in bedrock and mines.

Bedrock breaks down to form soil. During the process, some of the lead of the parent rock remains in the soil and some is dissolved in groundwater which may eventually enter into streams. Part of the lead results from the radioactive decay of radon gas. It, therefore, may be expected to have a very different distribution from dissolved lead inasmuch as part of it escapes into the atmosphere and later returns as natural fallout in rain.

Like several trace elements lead becomes concentrated in the organic material which forms coal. Thus when coal is mined and burned lead may become available to man by a direct transformation of the bedrock. That this may be a potent source of lead is indicated by results of the "Tri City Lead Study" which indicated much higher atmospheric lead concentrations in the winter in Chicago than could be attributed to automobile exhausts. Lacking a positive means of identifying lead from different sources the author of this study could only

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COSMIC AND RADIOGENIC

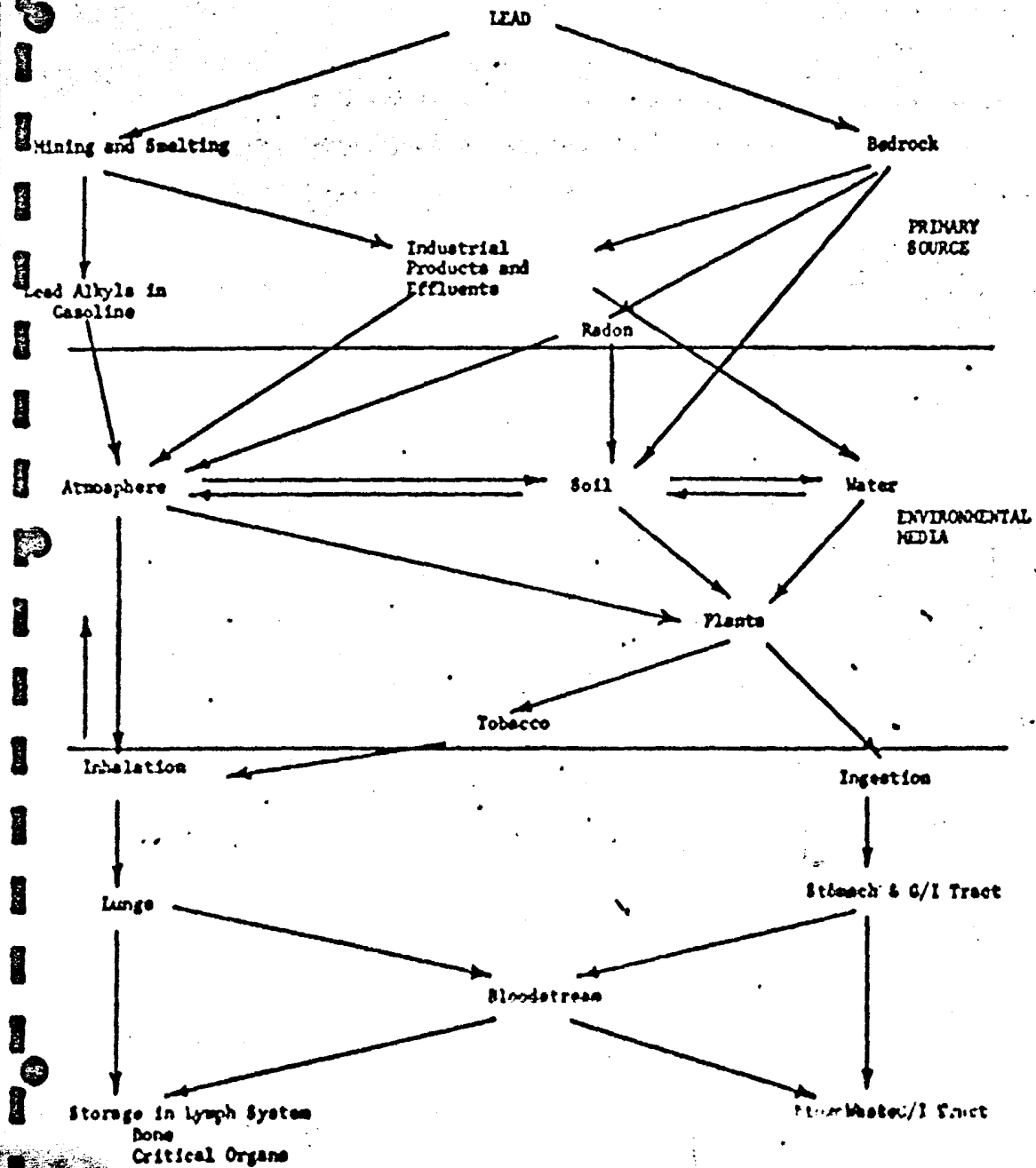


FIGURE 1.

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speculate that the lead originated in coal fly ash. Because geologic processes leading to the formation of coal are very different from those leading to hydrothermal ore deposits, identification of lead from these different sources by isotopic characterization may be possible.

Most of the lead in man's present environment comes from mines. It enters the environment as various industrial products ranging from storage batteries to insecticides and lead alkyls in gasolines. If all of the lead in these various products were in fact available to man there would no doubt be great cause for concern. However, much of the lead in lead products enters the environment in inert forms. It is known for example, that much of the lead from automobile exhausts is found in relatively large particles which fall out rapidly and become part of the soil. As long as food is imported from country areas the level of lead in roadside soils is not likely to be an important environmental health factor.

On the other hand, material in the submicron range may stay in the air for extended periods, be breathed in and have a high probability of absorption in the body. Thus there may be available and non-available fractions of the same material. In general, lead which persists in the atmosphere for some time, or finds its way into soil and water is most likely to be available to plants and to man.

Not all lead which is available for entry into man is harmful because it may enter in a form which is absorbed with low efficiency. This lead, although available to man in a sense, is not available for entry into important biological processes. By isotopic characterization of lead in biological samples it may be possible to identify the sources of lead biologically available to man. For example, if the lead ingested from plant material is largely unabsorbed

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and is eliminated in the feces it may be isotopically distinct from airborne lead which is absorbed through the lungs and shows up in the urine or is accumulated in the bones or teeth.

The use of lead isotopic characterization is a means of identifying the geochemical and biochemical pathways followed by lead to and through man, depends on the existence of natural isotopic variations. The extent to which useful isotopic distinctions exist and the principles which explain their origin are discussed in the following section.

The Origin of Isotopic Variations in Lead

Variations in isotopic abundances occur widely in nature and in general stem from two distinct mechanisms.

The first concerns the thermodynamical properties of isotopic compounds in chemical, physicochemical or biochemical process. Since the reaction kinetics are mass dependent, a lighter isotope will react more favorably than a heavier one in the same reaction. The differences are more marked in the lighter elements, in particular in hydrogen, carbon, nitrogen and oxygen, the biological tissue building elements.

The second source of isotopic abundance variation occurs where an existing isotope is mixed with a daughter product of a radioactive decay series. Potassium-40, for instance, decays to argon-40 and changes the argon 36:38:40 ratio with time. Of particular interest to the present discussion is the contribution to the world's lead inventory by the decay of the unstable elements uranium and thorium.

Common lead is found to comprise a mixture of four stable isotopes,

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Pb-204, Pb-206, Pb-207, and Pb-208. There are also as many as sixteen different radioactive lead isotopes known, but the principal one pertinent to this discussion is Pb-210, also known as Radium D.

Lead-204 is not known to have been formed by any natural decay or any other nuclear reaction. It, thus, constitutes a measure of the original lead which was formed when the elements were formed, sometimes known as primaeval lead. The isotopic ratio of Common Lead mined today results from the addition to primaeval lead of radiogenic lead, Pb-206, Pb-207, and Pb-208, formed from the decay of uranium and thorium. Figure (2), (3), and (4) show the decay schemes for the three radioactive families resulting in the stable isotopes of lead. The half-lives of the parents, U-238, U-235, and Th-232, being the longest in each series, determine the ultimate rates of buildup of the radiogenic isotopes. In the earth's crust as a whole, Pb-206 and Pb-208 are being produced at about the same rate at the present time, while Pb-207 is being produced at a much slower rate because of the low abundance of U-235. In early geological time, however, when U-235 was much more abundant, Pb-207 was being formed more rapidly than Pb-206.

The differences in the isotopic ratios of common lead ore may be, therefore, attributable to the manner of the addition of the radiogenic lead to the primaeval lead.

1. The ores may have been separated from their sources at different times. Recently separated ores can be expected to contain greater quantities of radiogenic lead than ores separated earlier.
2. The abundance of each radioactive parent may have been variable.
3. The intermediate daughter nuclides preceding lead in the three decay schemes may have been differently dispersed prior to becoming lead.



FIGURE 2.

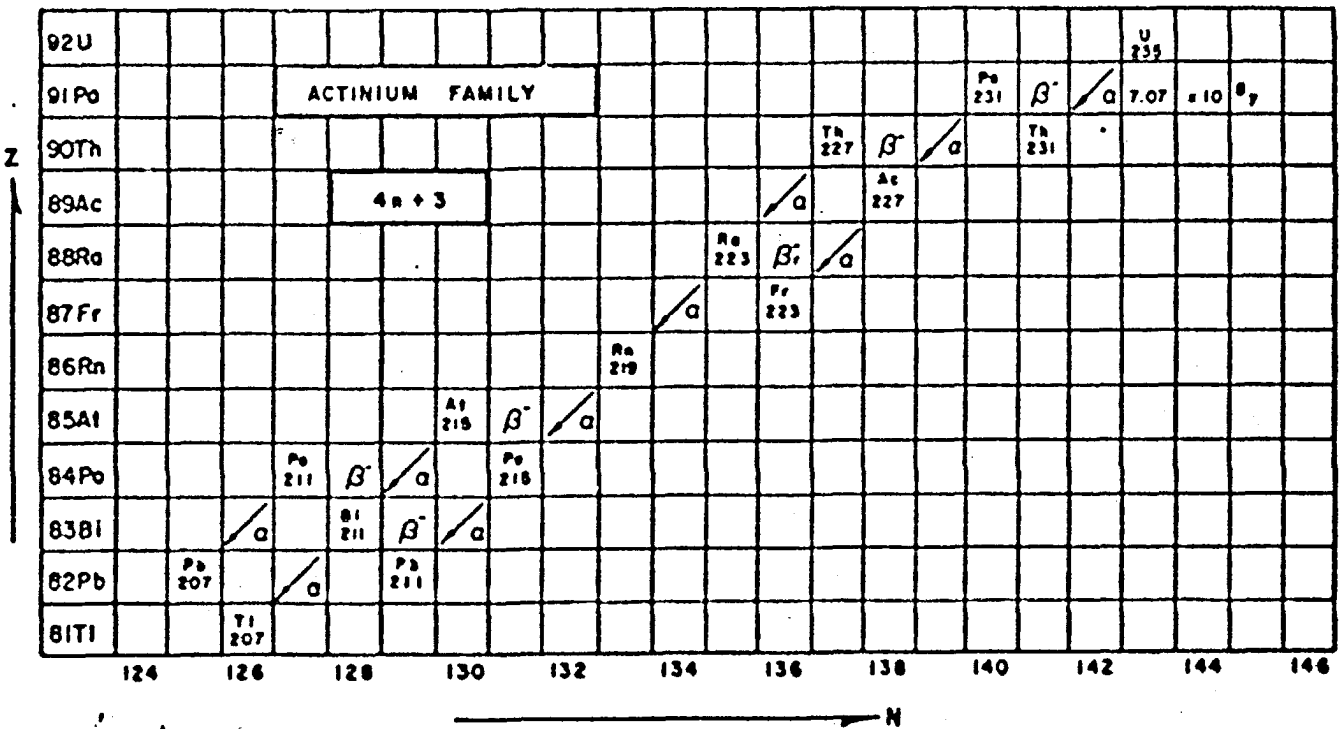


FIGURE 3.

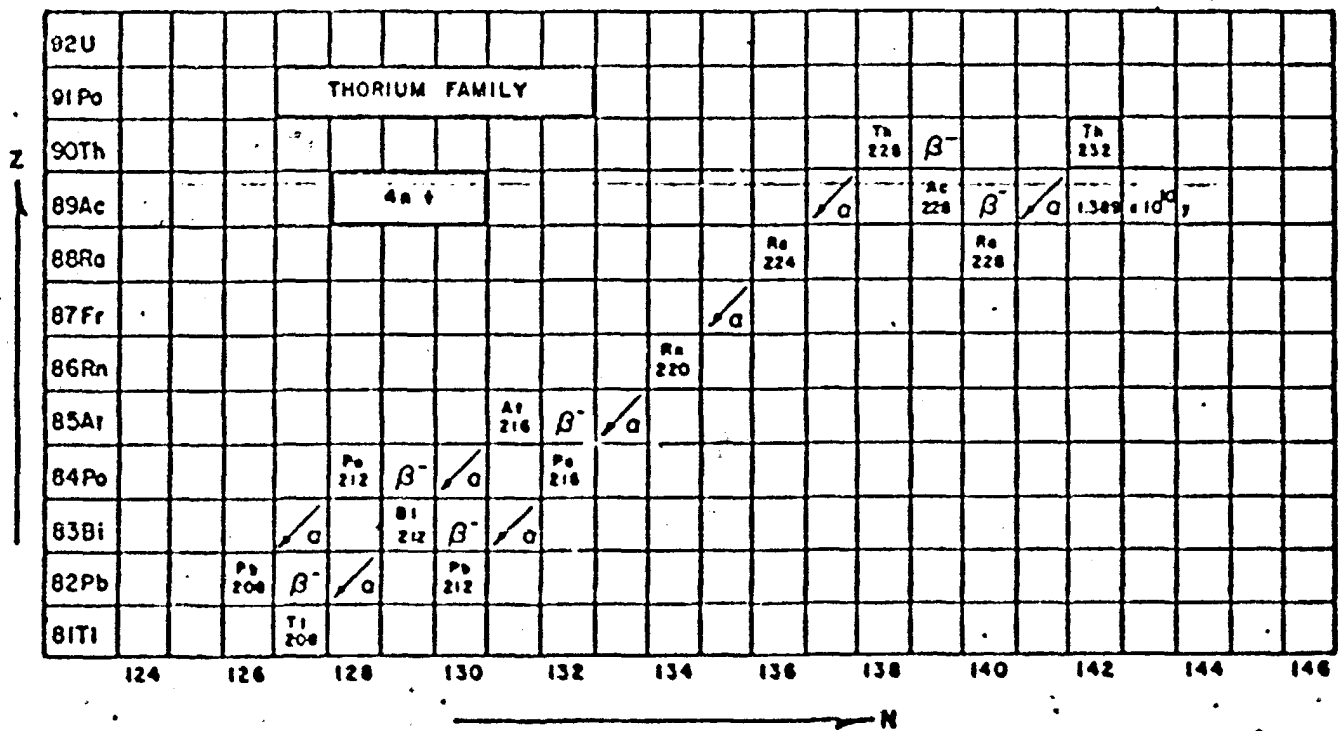


FIGURE 4.

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4. The foregoing causes may become combined, in any proportion, to make new ore bodies in a variety of geological processes.

The existence of these isotopic abundance ratios has been widely used to determine the history of lead ore deposits, mostly galenas, and much has been published discussing the possible interpretation of the considerable number of measurements in the literature. It is, however, the fact, and the characteristics of, these variations in major industrial lead ore areas that are pertinent to the present discussion. One of the prime objectives of the proposed study is to differentiate in man and his environment between lead derived from local natural sources and that from distant areas via industrial sources and pathways. To achieve this, it must first be demonstrated that the isotopic ratios of lead in local soils and groundwaters can be distinguished from lead derived from the major industrial areas. This would appear to be clearly possible in certain areas. In others the range from the separate sources may overlap such that definitive results are impossible. It is the primary purpose of the initial investigation under this project to define the extent to which the identification of specific sources is possible in the present urban scene.

An additional parameter which may prove to be a valuable tool to characterize lead in man and his environment is the specific activity of the radioactive Pb-210. Leads associated with little residual U-238, such as industrial lead, would contain little Pb-210. Soils and groundwaters on the other hand should contain considerable quantities derived from the U-238 in the weathered bedrock. This could be a useful indicator of whether or not the lead in plants is soil or atmosphere derived.

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Natural Variations of Lead Isotope Composition

As a result of the factors discussed above, significant variation in lead isotope abundances are observed in lead sources from different geochemical backgrounds. Table I shows a selected list of lead samples from various mining districts to emphasize the great range in composition. Also given is the value for "primeval" lead as derived from a uranium-free metallic meteorite. The lead in the manganese nodule from the ocean floor probably represents the average composition of soluble lead that entered the Pacific Ocean over the past few hundred years. In each geographical area the geology of the surface will determine the lead isotope composition in the drinking water and in food grown in the soil. The isotopic variations in soils over the world will vary as widely as that in samples of lead listed in Table I.

The indicated experimental error shows that the natural variations are large compared with the analytical capability. Some mines or mining districts have rather uniform isotopic composition. Examples are Broken Hill, Coeur d'Alene and Balmat. On the other hand, wide variations are observed in the Mississippi Valley type ores such as those of Southeast Missouri District and the Tri-State District in western Missouri. Nevertheless, the average lead isotope composition from these areas is reasonably uniform when integrated over weeks of mining.

Variations in the isotopic composition of lead produced by the lead alkyl manufacturers would relate to the original sources of lead in the pool, changes in the ratios of these sources and the method of warehousing the raw material. Some estimate of these variations can be made from the records of procurement and handling, but better values can probably be obtained by analyses of production on a periodic basis.

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In the New York City area, for example, an initial survey should include the isotopic composition of lead in: drinking water, average diet (perhaps from routine A.E.C. Sr^{90} samples), lead in gasoline (sample major depots), lead in Consolidated Edison coal, and average lead in city air. It would appear from Table I that identification of these sources in the human lead burden may be feasible.

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TABLE I

Isotopic Composition of Selected List of Lead Samples

	Pb^{206}/Pb^{204}	Pb^{207}/Pb^{204}	Pb^{208}/Pb^{204}
Canon Diablo (meteorite)	9.5	10.3	29.4
So. Africa (Rosetta Mine)	12.6	14.1	32.8
North West Terr. (Con Mine)	14.1	15.1	33.9
Cobalt, Ontario	14.9	15.3	34.8
Broken Hill, N. SW.	16.2	15.6	35.9
Coeur d'Alene, Idaho (Sullivan Mine, B.C.)	16.5	15.6	36.8
Balmat, N.Y.	16.9	15.6	36.9
Franklin, N.J.	17.4	15.7	37.7
Montana (Boulder Batholith Mines)	18.1	15.6	38.5
Cerro de Pasco, Peru	18.8	15.7	39.0
Mascot, Tenn.	19.6	15.8	39.7
S.E. Missouri (Av.)	20.5	16.0	39.9
Tri State (Joplin) (Av.)	22.2	16.1	41.9
Manganese Nodule (Ocean Floor)	18.9	15.7	38.7
Experimental Error	$\pm .05$	$\pm .05$	± 0.1

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III. SCOPE OF WORK

The proposed study of the use of lead isotopic composition as an environmental tracer will be carried out in three phases. Phase I will be concerned with the development and evaluation of analytical techniques. In Phase II the feasibility of using isotopic composition to characterize the lead in primary sources, environmental media, and in man will be established, and the use of tree rings as paleo-environmental indicators will be investigated. Phase III will extend the techniques and principles established in previous phases to several different geographical locations in order to prove the generality of their application.

Phase I. Analytical Technique Development and Evaluation

The unique aspect of the proposed approach to the study of environmental lead is the use of stable and radioactive isotopes to characterize the lead from various environmental sources. Although much experience has been gained, (to a large extent by Isotopes, Inc. personnel) in measuring lead isotope ratios, in measuring low concentration of lead by isotope dilution, and in measuring low levels of Pb-210 radioactivity, the problems associated with the ultra-microanalysis required for environmental samples must be approached with care and thoroughness not commonly exercised in routine trace analysis. The first six months of work, Phase I, will, therefore, be concerned with construction of a lead-free laboratory, production and testing of lead-free reagents, preparation of standards, and establishing the limits of detection of the methods used.

The precision and accuracy obtainable for both the abundance and isotopic measurements will be defined. Intercalibration will be conducted between the lead abundance measurements done at Isotopes, Inc. by the isotope dilution-mass spectrometer method and those done by laboratories using the dithionite-colorimetric technique.

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Procedures for measurement of Pb-210 in low level samples will be established and evaluated and the use of Pb-210 for yield determination in chemical concentration procedures will be developed with particular regard to the sample matrices.

Study of typical environmental samples will be done to check the procedures. Isotopic analysis of lead alloys from tetraethyl-lead manufacturers will be undertaken to establish the range of isotopic variability to be expected in leaded gasolines.

Throughout Phase I a continuing effort will be devoted to studying the problem of environmental lead in broad perspective and toward the end of this time attention will be directed toward design of critical experiments to be performed in Phase II.

Phase II. Feasibility Study of Isotopic Characterization of Lead in Primary Sources, Environmental Media, and in Man

Following the thorough development and evaluation of lead isotope measurement techniques for environmental samples in Phase I, pilot studies will be conducted to test the feasibility of using isotopic characterization as a means of distinguishing various sources of environmental lead. As a result of experimental design studies undertaken in Phase I, at least two areas in the New York City area will be chosen for environmental sampling.

An attempt will be made to identify and characterize primary sources of lead such as bedrock and soils, coal and fly-ash, lead alkyls in gasoline, and tobacco. Background information on the samples analysed will be studied in order to understand the origin of the isotopic composition observed.

In general the lead in primary sources is not available to man. It must first enter into environmental media such as air, water, and food plants to become a biological contaminant of man. The sources of environmental lead

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available to man will, therefore, be sampled and compared with the primary sources in the same area to determine which primary sources are responsible for the available environmental lead.

A third aspect of the pilot studies will include isotopic characterization of human samples including bone, teeth, blood, and urine to determine the relation between available environmental lead and that actually found in man himself. In both the selection of sample type and procurement of samples of human material guidance and cooperation will be sought from experts in lead biochemistry and toxicology.

To place the results of the environmental sampling program in historical perspective it is important to establish the feasibility of using tree rings, soil profiles, and sedimentary layers as records of changing levels of environmental lead. Lead isotope studies of tree rings are particularly important because relatively high lead concentration in outer tree rings has been used as prima facie evidence of severe environmental contamination. The validity of these conclusions cannot be accepted unless it can be shown that the lead in the zones of high concentration is isotopically different from the lead in the bedrock and soil and the same as that in the atmosphere. Our measurements will be capable of establishing these important relationships.

It is anticipated that correlative environmental studies will be underway during the time period of Phase II. Exchange of information with these programs and isotopic analysis of significant samples will enhance the value of the whole research project. Because the isotopic tracer technique may be especially valuable in investigations of lead metabolism in biological systems close attention will be given to the development of cooperative research programs.

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Phase III. Extended Environmental Studies.

Once the analytical methods for lead isotopic characterization and the feasibility of their application are well established, study of different geographical locations will be undertaken. Areas with different geology, meteorology and industrial activity will be studied using the New York City pilot studies as models.

During this phase continued and increased participation and cooperation with other groups and agencies is anticipated. The specific scope of work to be undertaken in Phase III cannot be defined at this time because it is strongly dependent on the results of the feasibility study in Phase II and the extent and nature of the correlative study programs of other groups.

IV. EFFORT AND COST

Isotopes, Inc. regards the proposed program as a basic research effort which will require the utilization of uniquely qualified personnel. The program must also have some expensive and special facilities at its disposal. Isotopes, Inc. will provide both of these requirements within the basic cost of the project scientists.

The project scientists will have office space, clean chemical laboratory with special hoods, solid source mass spectrometer, emission spectrograph, electron diffraction and electron micrograph at their disposal. They will have all necessary secretarial, drafting and technical support. In the technical area this will include electronic support, data processing, machine shop effort and chemical technician labor. The basic costs also include the continuing advisory support of Drs. Kulp, Willis, Friend and Bate as described above.

The work is divided into three phases, each of six months duration. Funding may therefore be done in six, twelve or eighteen month increments. It is understood that quarterly technical reviews and six months technical reports will be part of Isotopes, Inc. commitment. Further, it is agreed that since the customer is primarily buying "a level of effort" that the scope of work may be modified as a result of the Quarterly Reviews. Particularly, if is expected that redefinition and elaboration of the scope of work for Phases II and III will occur prior to the initiation of Phase II.

In Phase I no direct charges are involved except travel and subsistence assuming two trips to the Midwest. In Phase II, an allowance is made for additional travel involved in local sampling. In Phase III, it is

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anticipated that there will be some increased travel and a requirement for some special sampling gear. In all cases the direct charges will be billed as actual plus an administrative charge of 10%.

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Phase I - July 1, 1966 - December 31, 1966 (six months)

<u>Technical Effort</u>	<u>Project Cost</u> <u>Rate per Month Incl. All Support</u>	<u>Total</u>
Scientist-Project Director (6 mos.)	\$5080.	\$30,480
Associate Scientist (5 mos.)	3720.	18,600
Chemist (3 mos.)	2200.	6,600
		<u>55,680</u>

Direct Charges

Travel and Subsistence		
2 Trips to Mid West, total 8 days		Est. 500
	Total	<u>\$56,180</u>

Phase II - January 1, 1967 - June 30, 1967 (six months)

<u>Technical Effort</u>	<u>Project Cost</u> <u>Rate per Month Incl. All Support</u>	<u>Total</u>
Scientist-Project Director (6 mos.)	5080.	30,480
Associate Scientist (6 mos.)	3720.	22,320
Chemist (6 mos.)	2200.	13,200
		<u>66,000</u>

Direct Charges

Travel and Subsistence		Est. 1,000
	Total	<u>\$67,000</u>

Phase III - July 1, 1967 - December 31, 1967 (six months)

<u>Technical Effort</u>	<u>Project Cost</u> <u>Rate per Month Incl. All Support</u>	<u>Total</u>
Scientist-Project Director (6 mos.)	5080.	30,480
Associate Scientist (6 mos.)	3720.	22,320
Chemist (6 mos.)	2200.	13,200
Field Samples (4 mos.)	2200.	8,800
		<u>74,800</u>

Direct Charges

Travel and Subsistence		Est. 2,000
Special Sampling Gear		5,000
	Total	<u>\$81,800</u>

V. PROJECT PERSONNEL

The proposed analytical methods research project will be conducted by a special project group established within the technical organization.

Dr. Donald F. Schutz will serve as the Project Director and principal investigator. Dr. Schutz is a geochemist on the Research Department staff and is currently responsible for the Nuclear Operations Section of that Department. He is highly qualified to direct the proposed lead isotopic analysis research by virtue of his extensive experience in the development of a variety of analytical methods for determining trace element concentrations in seawater. As Project Director, Dr. Schutz will be responsible for the scientific direction and management aspects of the research studies. In this position he will report to Dr. C. E. Crouthamel, Director of Research of Isotopes, Inc.

Several senior scientists in Isotopes, Inc. who have unique backgrounds from which to contribute to this program will advise Dr. Schutz in the direction of this project. These include: Dr. J. Laurence Kulp who has directed major research programs in lead isotope geochemistry, mass spectrometric development, worldwide radioactive fallout and the movement of strontium-90 in the human food chain; Dr. James P. Friend, a physical chemist with extensive experience in the field of atmospheric aerosols and particle studies; Dr. George L. Bate who has conducted numerous research studies involving isotopic lead determinations and lead geochronometry; and Dr. Eric H. Willis who is experienced in research work based on mass spectrometer measurements of stable isotope ratios and low level radiological determinations of carbon-14 and other radioisotopes.

Mr. Roland Kulogrivov, head of the Stable Isotopes Analysis Section of the Scientific Services Department, will serve as the physical analyst for

Isotopes, Inc.

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this project. He will be primarily responsible for the performance of mass spectrometric analyses and the development of suitable new analysis techniques.

Dr. J. Marion Waspler of the Georgia Institute of Technology Earth Sciences Department, who has done significant work in isotopic analysis of lead in very low concentration, will participate in the program in the summer of 1966 and will continue as an advisor in analytical techniques.

One qualified technician will be assigned to the project group to assist in sample preparation and analysis and to perform initial data reduction calculations.

Detailed resumes of the professional qualifications and experience of the scientists assigned to the proposed project group are included at the end of this section. Publication lists are also included and have been annotated with asterisks (***) to indicate those papers which are especially relevant to the lead isotope analysis studies.



RESUME

GEORGE L. BATE, Ph. D.

Scientist

Dr. Bate serves on the Research Staff at Isotopes, Inc. as a specialist in nuclear physics. As Associate Project Director of Project PINOCCHIO, he has over-all scientific and administrative responsibility for the coordination of the research studies for the project and participates in specific phases of the research when necessary. Project studies are in the areas of nuclear parameters, geology, meteorology, gas analysis techniques, and local air analyses. He is also conducting theoretical studies of the potential usefulness of radionuclides formed by fast neutron spallation reactions and heavy particle reactions which accompany the detonation of nuclear devices in various geologic media. Dr. Bate previously directed a project to establish and evaluate methods for the detection of ultra-low-level radioactive gaseous effluents from nuclear operations. He has additional responsibilities on the Patents, Radiation Safety, and Technical Studies Committees of the company.

Before coming to Isotopes, Inc. in 1965, Dr. Bate taught advanced physics courses as a Professor of Physics at Wheaton College and was responsible for the installation of a radioisotope laboratory there.

From 1956 to 1965 Dr. Bate served as a part-time Research Consultant and Associate at the Argonne National Laboratory. His work there produced the first definitive assay of thorium in both stone and iron meteorites and led to further definition of the abundances of certain other elements in cosmic and terrestrial sources. Later research permitted further development of models of nuclear fission by evaluation of excitation functions and angular distribution of fragments of nuclear fission induced by charged particle bombardment.

As a Research Assistant at the Lamont Geological Observatory of Columbia University from 1951 to 1954, Dr. Bate performed research which led to a determination of the age of the earth.

Dr. Bate received a B.A. in Physics from Princeton University in 1945 and an M.S. in Physics from the California Institute of Technology in 1946. He was awarded a Ph. D. in Nuclear Geophysics from Columbia University in 1955. Dr. Bate is a member of Phi Beta Kappa and Sigma Xi. He holds memberships in the American Association of Physics Teachers, the American Association for the Advancement of Science, and the Geochemical Society.



PUBLICATIONS

GEORGE L. BATE

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- *** J. Laurence Kulp, George L. Bate and Bruno J. Giletti, "New Age Determinations by the Lead Method", Proc. Geol. Assoc. Can. 7, 15-24, May 1955
- *** J. Laurence Kulp, George L. Bate and Wallace S. Broecker, "Present Status of the Lead Method of Age Determination", Am. Jour. Science, 252, 345-365, June 1954
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CARL E. CROUTHAMEL

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- C. E. Crouthamel and H. Diehl, "Gas Analysis Apparatus Employing the Velocity of Sound", Anal. Chem. 20, 515, June 1948



RESUME

Carl E. Crouthamel, Ph. D.
Director of Research

As the Director of the Research Department at Isotopes, Inc., Dr. Crouthamel directs the contract and in-house research activities of the company. His staff includes experts in a variety of scientific disciplines including nuclear and radiochemistry, geochemistry, geology, oceanography, biochemistry and nuclear physics.

Dr. Crouthamel was employed at the Argonne National Laboratory as a Senior Chemist from 1950 to 1965. He initially developed methods for the analysis of reactor fuel and was responsible for the organization and supervision of the radiochemical program in the Chemical Engineering Division. For twelve years he was the Group Leader of the Reactor Chemistry Group, with responsibility for the determination of reactor materials cross sections, development of activation methods associated with fast breeder reactors, and development and application of such sophisticated counting techniques as gas proportional, scintillation, and solid state spectrometers. He is well-known for his pioneering work on the development and application of sodium iodide-thallium activated scintillation spectrometers and multi-channel analyzer systems.

During the past five years he has been primarily engaged in studies of fused salt-liquid metal systems and their application to the direct conversion of heat to electricity. These activities included extensive basic research in fused salt and intermetallic systems with particular emphasis on the solubility of the lithium halides and sodium halides in low-dielectric covalent solvents such as fused lithium hydride and anhydrous acetic acid. In addition he has investigated the thermodynamic properties of bimetallic systems, fused salt binary and ternary systems, and mixed intermetallic fused salt systems. He also designed an inert helium atmosphere box as a basic research unit which has been widely copied and reproduced at Argonne as well as many other laboratories.

Dr. Crouthamel received a B.S. in Chemistry and Mathematics in 1942 from Eastern Nazarene College and his M.A. in Analytical Chemistry in 1943 from Boston University. Doctoral research was done at Iowa State University and he obtained his Ph.D. in Inorganic Chemistry in 1950. He has been author of more than 42 papers in his fields of investigation and has edited and contributed substantially to four books. Dr. Crouthamel is a member of the American Chemical Society, the American Nuclear Society, the Research Society of America, the New York Academy of Science, and the American Association for the Advancement of Science.



RESUME

JAMES P. FRIEND, Ph. D.
Senior Scientist

Dr. Friend is a physical chemistry specialist on the Research Department staff and also acts as the Senior Scientific Advisor for the Department. He is presently conducting detection probability studies and field experiments under Project PINOCCHIO for the VELA On-Site Inspection Program. He is also engaged in research in the field of atmospheric chemistry. This work includes studies of the composition and characteristics of stratospheric aerosols and the relationship of radioactivity to particulate matter in the atmosphere. In addition, Dr. Friend is directing a study of the mechanisms of transport in the stratosphere and deposition on the ground of radioactive fallout.

Dr. Friend served as Director of Project STAR DUST for Isotopes, Inc. In this capacity he coordinated the efforts of meteorologists, interpretive scientists and analysts. Under the High Altitude Sampling Program, he studied fluid flow in air sampling devices and worked on the calibration of the WU-2 sampling ducts.

Other experience at Isotopes, Inc. has included studies of the physical and chemical properties of environmental particulate materials and aerosols, establishing procedures for sampling of particulate matter in the atmosphere and in soils; and preparation of procedures for plutonium analyses and interpretation of these analyses. He was also co-author of the AEC report "Study of the General Feasibility of Radioisotopes Methods in the Natural Gas Industry". He has supervised the analytical efforts in the gas counting laboratory where carbon-14, hydrogen-3, radium-226 and radon measurements are made.

Prior to his employment at Isotopes, Inc., Dr. Friend was employed as a Physical Chemist at the Perkin-Elmer Corporation where he performed gas chromatographic investigations.

During his military service, Dr. Friend was assigned to the Army Chemical Center where his work involved the development of infrared instrumentation and the detection of chemical warfare agents in the atmosphere.

Dr. Friend received both his M.A. (1953) and Ph. D. (1956) in Physical Chemistry from Columbia University. His thesis subject was microwave spectroscopy. He received a B.S. in Chemistry from Massachusetts Institute of Technology and is a member of the following professional societies: American Physical Society, American Association for the Advancement of Science, and Sigma Xi.



PUBLICATIONS

JAMES P. FRIEND

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*** J. P. Friend, "Properties of the Stratospheric Aerosol", Tellus 1966 (In Press)



RESUME

ROLAND KOLOGRIVOV, B.S.

Senior Associate Scientist

Mr. Kologrivov has had six years experience in mass spectrometry particularly related to the isotope dilution measurement of rubidium, strontium, potassium, argon, uranium, thorium and lead. At Isotopes, Inc. he has supervised the installation of the Reynolds type mass spectrometer used for gas measurements and the Shields type (CEC-21-703) instrument used for solid source measurements. He designed and supervised the construction of the high vacuum fusion system used to prepare samples and calibrate the isotopic tracers used in the isotope dilution method. These systems utilize the most recent developments in the state-of-the-art of ultra high vacuum techniques oriented to geochemical age determinations. His present work includes supervision of the mass spectrometry laboratories.

Mr. Kologrivov was previously employed at the Lamont Geological Observatory of Columbia University where he was in charge of the potassium-argon dating laboratory. These analytical techniques also utilized high sensitivity gas mass spectrometry and ultra high vacuum techniques. Other work at Columbia University included low level detection of radium in human bone and developmental work on low level nuclear detectors and counting techniques.

Mr. Kologrivov received a B.S. in Chemistry from City College of New York and is a member of the American Vacuum Society, the New York Academy of Sciences, the American Nuclear Society, and the American Association for the Advancement of Science.



PUBLICATIONS

R. KOLOGRIVOV

A. Walton, R. Kologrivov and J. L. Kulp, "The Concentration and Distribution of Radium in the Normal Human Skeleton", Health Physics 1, 409-416, 1959

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RESUME

J. LAURENCE KULP, Ph.D.

President

Dr. Kulp has directed a wide variety of research projects in the general areas of geochemistry and radiochemistry. These have included various aspects of the dissemination of radioactive debris into the geosphere and biosphere. Major projects have been Projects HASP and STAR DUST (world-wide radioactive fallout in the atmosphere), biogeochemistry of Sr^{90} and Cs^{137} , movement of bomb-produced C^{14} in the atmosphere, biosphere and oceans, mathematical model for the prediction of fission products in post-attack diets, decontamination of aircraft from fission debris, plutonium hazards from non-nuclear accidental destruction of devices and Project Vela Cloud Cap.

Geochronology has also been a focus of study. Investigations have been made on the development of the method of radiocarbon dating and its application to archeological and geological problems in such diverse areas as the constancy of cosmic ray flux, the origin of petroleum and the development of the human culture in South America. The K-Ar, Rb-Sr and U-Pb methods of dating rocks have been developed in Dr. Kulp's laboratory and applied to numerous geological problems. In 1962 a definitive geological time scale was produced.

Other fundamental investigations have been in the field of isotope geochemistry as applied to the origin of the Mississippi Valley lead ore, of the porphyry copper ore of the Southwest, of salt dome sulfur deposits, the uranium deposits of the Colorado Plateau and the age of the gold ore of Lead, S.D. Studies utilizing lead and strontium isotope variations have helped shed light on certain problems of the evolution of the continents and ocean basins.

In space science, Dr. Kulp has been concerned with the geologic mapping of the moon by gamma spectrometry, the age of meteorites, the probable sources of water on the lunar surface and the radiation background to be expected in cis-lunar space.

From 1947 until 1965, Dr. Kulp was on the faculty of Columbia University, as Professor of Geochemistry. From 1954 until 1965 he was Director of the Geochemistry Laboratory at the Lamont Geological Observatory. He is presently associated with Columbia as an Adjunct Professor. He has served as a consultant to industrial firms, military groups, and other Government agencies, i.e. Esso Standard Oil, Humble Oil, Kennecott Copper, Gulf Research and Development, Rand Corporation, U.S.A.F., Army Ordnance Corps, U.S.A.F.C., Federal Government of West Germany.

Dr. Kulp was a founding Director of Isotopes, Inc., and served in a consulting capacity as Senior Scientific Advisor until December, 1964 when he became President of the company. He is a member or fellow of the following scientific societies: American Chemical Society, American Physical Society, Geological Society of America, Geochemical Society, Mineralogical Society of America, Association of Petroleum Geologists, American Geophysical Union, Society of Sigma Xi, American Association for the Advancement of Science.



PUBLICATIONS

J. L. KULP

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- W.S. Broecker and J.L. Kulp, "The Radiocarbon Method of Age Determination", *American Antiquity* 22, 1-11 1956
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RESUME

DONALD F. SCHUTZ, Ph. D.
Scientist

Dr. Schutz is a member of the scientific staff in the Research Department at Isotopes, Inc. In this position he provides extensive experience and capability to the research staff in the fields of geochemistry and oceanography. He has recently been assigned as Manager of the Nuclear Operations Section of the Research Department to exercise over-all technical and administrative responsibility for projects involving research and development of nuclear test detection and on-site inspection techniques. Dr. Schutz is uniquely qualified for this coordinating responsibility because of his previous experience as the Director of Project PINOCCHIO where he supervised the development of radiological techniques for the detection of clandestine nuclear weapons tests. He also brings a valuable geological and oceanographic background to this position which requires a broad knowledge of environmental factors and their influence on nuclear operations projects. His earlier work at Isotopes, Inc. included the development of improved techniques for extracting inert gases from air samples, and the evaluation of internal gas proportional counting methods for radiometric assay of separated gases. In addition to his laboratory studies on these problems, Dr. Schutz also participated in and directed field operations in connection with several contract research programs.

Before coming to Isotopes, Inc., Dr. Schutz was a Research Staff Geologist in the Department of Geology at Yale University. His work there was primarily concerned with the development of analytical techniques for the determinations of trace elements in seawater by neutron activation and X-ray fluorescence analysis. In a study of the geochemical problems of marine trace element economy and distribution, these methods were applied to a worldwide sampling of seawater. The samples included those collected by Dr. Schutz in the Antarctic during the winter of 1963-64 on the oceanographic research vessel ELTANIN. He has also studied the distribution of silver in Connecticut streams and the Long Island Sound by emission spectrographic analysis in an investigation of the effect of industrial processes on the trace element concentration of river water.

At Rice University Dr. Schutz completed a statistical study of regional variations in the chemical composition of basaltic rocks. During the summers of 1955 and 1956 he worked for Bear Creek Mining Company on geochemical prospecting projects in the states of Maine and Arizona.

Dr. Schutz received his Ph. D. in Geology in 1964 from Yale University. His thesis was concerned with the geographical and vertical distribution of over eighteen trace elements in seawater. He received an M.A. in Geology from Rice University in 1954 and a B.S. in Geology in 1956 from Yale University. He is a member of Sigma Xi, the Geochemical Society and the American Geophysical Union.



PUBLICATIONS

DONALD F. SCHUTZ

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RESUME

ERIC H. WILLIS, Ph. D.
Senior Research Scientist

Dr. Willis is Manager of the Scientific Services Department at Isotopes, Inc. He has responsibility for the Mass Spectrometry, Gas Analysis, Radiochemistry, Radioassay and Solid Phase Analysis activities of the company. A significant portion of the analytical research and routine sample analysis work under his direction is conducted in support of Projects MOCCASIN and PINOCCHIO of the VELA ON-SITE Inspection Program. In addition, Dr. Willis personally participated in Project Pre-BRECCIA sampling and analysis in connection with Operation LONG SHOT at Amchitka Island, Alaska.

Prior to joining Isotopes, Inc., Dr. Willis was an Assistant Director of Research in the University of Cambridge, England. He established the most advanced radiocarbon dating laboratory in the United Kingdom with wide applications in Quaternary Research, Soil Science and Archaeology. His geological investigations also include the study of palaeotemperatures in foraminiferal deposits using the relative abundance of the stable oxygen isotopes in a double collector isotope ratio mass spectrometer. The existence of artificial radiocarbon in the atmosphere due to thermonuclear weapon tests led him to study the activity levels at ground level and the lower stratosphere using high-flying aircraft of the R.A.F. in conjunction with A.W.R.E. Aldermaston. Being the most prolific of gaseous fallout products, the distribution of radiocarbon in the atmosphere with time yields considerable information concerning troposphere-stratosphere mixing times. The radioactive carbon dioxide was collected on molecular sieves in special wing-tip ducts and recovered by vacuum furnacing. The measurements were initiated shortly after the violation of the nuclear test moratorium in October, 1961 and are still continuing.

Dr. Willis introduced teaching courses in Isotopic Geochemistry at Cambridge University and lectured in Quaternary Stratigraphy. He was President of the Commission on the Absolute Age of Quaternary Deposits which convened as part of the International Quaternary Association in Denver, Colorado in 1965. At Cambridge he was a member of the committee directing meteorological studies of the University as well as supervising the control and housing of radioactive substances.

Earlier in his career Dr. Willis was employed by Nucleonic and Radiological Development Laboratories in chemical dosimetry under the auspices of the United Kingdom Atomic Energy Authority.

He received his Ph. D. in Radiochemistry from Cambridge University in 1956 and a B. Sc. in 1947 in Physics at Kings College, London.



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RESUME

J. MARION WAMPLER, Ph. D.

Consultant

Dr. Wampler is currently an Assistant Professor in the Earth Sciences Department of the Georgia Institute of Technology. From 1963 to 1965 he was a Research Associate at Brookhaven National Laboratory and in 1965 served as a Summer Research Participant at the Oak Ridge National Laboratory.

His research endeavors include isotopic studies of lead in sedimentary and metamorphic minerals, in tektites and related rock samples, and in archaeological objects and related lead ores. He has done uranium-lead age determinations and Mossbauer spectrometry of iron in clay minerals.

Dr. Wampler is co author of a number of articles dealing with isotopic composition and concentration of lead in some carbonate rocks, isotopic studies of lead in sedimentary pyrite, use of lead isotope analysis to determine the provenance of archaeological samples, and isotopic comparison of lead from Ivory Coast tektites and Bosumtwi Crater materials.

Dr. Wampler received his Ph. D. in Isotope Geochemistry from Columbia University in 1961; his B.A. in Chemistry from Bridgewater College in 1957. He is a member of the American Geophysical Union, the Geochemical Society, the American Association for the Advancement of Science and Sigma Xi.

Publications

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Isotopes, Inc.

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VI. FACILITIES AT ISOTOPES, INC. FOR PHYSICAL
AND RADIOMETRIC ANALYSIS

A. Mass Spectrometry

The mass spectrometric capability comprises three modern high precision instruments which cover a wide range of applications.

A Reynolds Gas Analysis instrument with a 4.5" radius tube and 60° sector focussing is equipped with ultra high vacuum techniques such that the ultimate detection sensitivity is 5×10^{-8} cc of argon.

A Shields type solid source mass spectrometer manufactured by Consolidated Electrodynamics has a 12" radius tube with 60° sector focussing. This is the principal instrument for lead abundance and isotopic ratio determinations. The over-all sensitivity of this machine for lead assay is summarized below:

Measurement	Precision	Sensitivity		
		As total lead	ppm solid samples	ppm liquid or soft tissue
Lead isotope ratios	High			
	$^{206}/^{207}$, $^{206}/^{208}$ to 0.2%	20 μ g definite	5	0.2 ppm
	$^{206}/^{204}$ to 0.4%	5 μ g probably with a little development	1	0.05 ppm
	Low			
	$^{206}/^{207}$, $^{206}/^{208}$ to 0.5%, $^{206}/^{204}$ to 1%	0.1 μ g	0.1	0.001 ppm
Lead abundance by isotope dilution	High (Approx. 5% of value)	0.002 μ g Assuming no blank	0.4*	0.004 ppm
	Low (Approx. 20% of value)	0.0005 μ g Assuming no blank	0.1*	0.001 ppm

*Could be lowered if very low reagent blank is achieved

Isotopes, Inc.

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The third instrument which completes the wide coverage of Isotopes, Inc. capability is a Nuclide Corporation RMS-15 spectrometer with 6" radius and 60° sector focussing with double beam collection. Isotopic differences of one part in ten thousand can be measured routinely.

B. Solid Particle Analysis

A filtered air clean room facility is equipped with two modern electron microscopes, a Hitachi HU-11B with resolution of 5 Å and a Philips EM 75 PXI with a 30 Å resolution. In addition an optical laboratory is equipped with a Leitz Ortholux Pol microscope and an American Optical Phase Contrast microscope.

C. Emission Spectrograph

Elemental analysis is performed on a Jarrell-Ash 3.5 meter Ebert-mount, research grating spectrograph.

D. Radioassay

Isotopes, Inc. offers a comprehensive range of instrumentation for the measurement of high, medium low and ultra low levels of radioactivity, details of which are given below.

Beta counters

(40) Low level -	background 0.2 - 0.6 cpm range - up to 500 cpm
(12) Medium level -	background 1.5 - 2.5 cpm range - up to 1000 cpm
(5) High level -	background 5 - 8 cpm range - 100 - 2000 cpm
(2) Automatic Sample Changer -	background 5 cpm range - up to 2000 cpm

2 Alpha-Beta Counters - background 1 cpm alpha, 60 cpm beta

Isotopes, Inc.

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Scintillation spectrometers

- (2) 3 x 3 NaI (Tl) Crystals, 50 Kev - 4 Mev gamma energies,
resolution 7.5%

"Well" NaI (Tl) Crystal, 50 Kev - 4.0 Mev, resolution 11.5%

- (2) X-ray Spectrometers, 5 Kev - 25 Kev, background - 1 cpm

Beta-Gamma Coincidence Spectrometer

γ energy - 10 Kev to 4.0 Mev

β energy - 0.3 Mev to 3.0 Mev

Backgrounds - 0.1 - 0.2 cpm

Liquid scintillation spectrometer (Automatic) - Packard Tri-Carb

- (4) Solid State Alpha Spectrometers

Backgrounds	.005 - .01
Resolutions	20 - 60 Kev
Range	entire alpha energy range

Total gamma counter

Background - 135 cpm
All gamma energies

- (2) X-ray proportional counters

Background - 1 cpm
Range - up to 1000 cpm

- (3) Alpha Scintillation Counters

Background - less than 0.4 cpm
Range - up to 6.0 Mev alpha particles

4 Absolute Calibration Counter

Background - 75 cpm

- (2) Ge(Li) - large volume gamma detectors

Resolution 4 - 8 Kev
Energy range - 30 Kev - 2 Mev

E. Lead-free Laboratory Facility

A new laboratory facility will be devoted to the project and will be furnished with lead-free surfaces and fittings. Teflon containers will be

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used where possible to avoid contamination of reagents. Critical work will be conducted in bench top hoods provided with dust-free air supplies.