

Isotopes of Uranium, Thorium and Lead, and Their Geophysical Significance

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In an effort to improve the data required in the calculation of geologic age a determination of the isotopes of the elements involved in the radioactive disintegrations (At. Nos. 81-92) was made by the magneto-optic technique of Allison. There appear to be four radioactive series beginning with eight isotopes of uranium (two to each) and ending with sixteen isotopes of lead (four to each). The relations of the constituent isotopes of uranium to

those of lead in a mineral are complex, and vary somewhat from mineral to mineral. Ordinary lead and radioderived lead possess different isotopic compositions though both apparently derive from the same ultimate source. The isotopic composition of lead changes with its age. The most accurate age determination would be obtained by confining the calculation to the U^{238} and Pb^{206} isotopes respectively.

THE most reliable of the several methods for the determination of geologic time is the one involving the ratio existing between the uranium occurring in any certain mineral and the lead produced from that uranium by the processes of radioactive disintegration. It is free from the great uncertainties associated with the other methods, and depends upon phenomena and facts which may be observed and tested within the laboratory. However, this method also has its own uncertainties. There is the possibility of the contamination of the radioderived lead by ordinary lead. In addition to this the almost universal presence of thorium introduces other probable errors. Obviously also the fundamental data are multiplied by a factor of 10^{11} (years) and it is highly desirable that they represent, as closely as possible, the actual substances which are represented by the formula. The chemical analysis of a mineral, upon which the ratio depends, gives all of the uranium and all of the lead, whereas the formula represents only a particular uranium and the particular lead derived from it. Could this particular uranium and lead be identified and estimated the data would then correspond with those theoretically implied by the formula. In order to accomplish this it was necessary to determine the various isotopes of uranium and of lead and to determine as far as possible their relative abundance.

Professor Aston's cooperation was secured and in 1927 he determined the principal isotopes of ordinary lead and in 1929 those of radioderived lead, using samples of lead tetramethyl pre-

pared for the purpose and supplied to him by Piggot.¹ These determinations settled several moot questions. They demonstrated that the accepted atomic weight 207.2 is a statistical mean of at least three isotopes (and probably more) and that one of these principal isotopes has a mass of 207 and is in all probability the end product of the actinium series. Furthermore, they demonstrated that radioderived lead has a different isotopic composition from ordinary lead. For, whereas in the latter the order of abundance of the respective isotopes is 208, 206, 207, in radioderived lead it is 206, 207, 208. The significance of these findings, from the point of view of the problem of geologic time, has been discussed by Piggot elsewhere.²

However, because of the rather wide limits of error which Aston assigned to his figures, and because, as yet, there was little known about the particular isotope of uranium from which the 206 lead came, the formula for the calculation of geologic age was not greatly refined. This aspect of the situation was discussed by Fenner in a letter to the Editor of Nature.³

There remained a pressing need for more knowledge of the isotopic composition of uranium and of lead, and in particular of samples of uranium and of the lead found associated with it in uranium minerals whose geologic and petrographic associations precluded, as far as possible, any contamination by ordinary lead; and which

¹ F. W. Aston, *Nature* **120**, 224 (1927); **123**, 313 (1929).

² C. S. Piggot, *J. Am. Chem. Soc.* **52**, 3161 (1930).

³ C. N. Fenner and C. S. Piggot, *Nature* **123**, 793 (1929).

also showed no signs of alteration, weathering, or enrichment of either of the principal constituents. This problem of the determination of the isotopes of uranium proved most baffling, despite the generous cooperation of Dr. Gomberg, Dr. Bainbridge, and others.⁴

Meanwhile Bishop and Lawrence, working in Professor Allison's laboratory, were studying lead by the magneto-optic technique, and had confirmed Aston's findings that the order of abundance of the three principal isotopes of ordinary lead is Pb^{208} , Pb^{206} , Pb^{207} , while in radio-derived lead the order of these three is Pb^{206} , Pb^{207} , Pb^{208} . In October, 1931 a cooperative undertaking was agreed upon between Dr. Allison and the writer which had as its objective the determination of the isotopes of all the elements from atomic No. 81 to atomic No. 92. This agreement, together with a continuation of investigations under way in Professor Allison's laboratory, has resulted in the determination of the isotopes of many of the radioactive elements and the correlation of the data so obtained into a system of radioactive interrelationships. This paper, and those accompanying it, constitute a report of the progress of this joint undertaking. The particular details appropriate to each subdivision of the problem are given in the individual papers but all the work was done with the essential unity of the investigation in mind. Each observation was repeated by several different observers and in many cases with more than one apparatus.

The findings have proved to be far more extensive and involved than was at first anticipated, and the explanation and interpretation of the data obtained have been a most difficult task to perform. These papers, therefore, seek to present the facts as we have obtained them, and to offer a suggested interpretation and scheme of the inter-related substances which does as little violence as possible to accepted and orthodox conceptions of radioactivity, in the hope that it may serve as a guide to the further elucidation of the many perplexing problems associated with the radioactive substances.

Samples for study were selected as follows: uranium from C.P. uranium chloride; thorium

⁴ Dr. A. C. Lane and his Committee on the Determination of Geologic Time, of the National Research Council.

from C.P. thorium chloride; radium from a standard radium solution; bismuth from C.P. bismuth chloride; lead from C.P. lead chloride; thallium from C.P. thallium chloride; uranium and its associated lead extracted from radium ore from Haut Katanga, Belgian Congo;⁵ uranium and its associated lead extracted from the pitchblende of the deposit at LaBine Point, Great Bear Lake, Canada;⁶ uranium and its associated lead from Swedish kolm. The kolm sample was collected by Professor A. H. Westergaard.⁷ The uranium and lead were obtained from the kolm by Dr. R. C. Wells in the course of his analysis of that material; lead extracted from Joachimsthal pitchblende residues.

The respective elements were weighed in the form of dry salts and added to distilled water⁸ to produce concentrations of the order of magnitude stated. When tested in the several magneto-optic apparatuses⁹ in Professor Allison's laboratory they gave the results and correlations shown in the accompanying tables and Figs. 2 and 3, which, we believe, demonstrate that uranium possesses eight isotopes, thorium eight, radium four, bismuth fourteen, lead sixteen, and thallium eight.¹⁰ The tables show the manner in which laboratory notes are taken by the observer using the lead data as an example, and indicate how the various isotopes "make their appearance," or "come in" as the concentration is increased by small increments, beginning with a dilution so attenuated that no minimum occurs, and continuing until one is just discernible to the observer.

An observation at which the observer discerns no effect at all is marked with a minus sign (—); a slight suggestion of a diminution of the intensity

⁵ The ore was as received at the Oolen plant of the Société Générale Métallurgique de Hoboken, and was obtained through the good offices of His Excellency, the Belgian Ambassador, Prince de Ligne, and F. Pisart of the Société Générale Métallurgique, Brussels.

⁶ By Dr. J. P. Marble while determining the atomic weight of this lead, and kindly supplied by him.

⁷ Who describes the location as "the middle and lower parts of the Peltura beds of the shale quarry at Gullhogen near Skovde in the province of Westergotland, Sweden."

⁸ The water was distilled first in an electric Barnstead still and then redistilled in an all-Pyrex still.

⁹ F. Allison, *Phys. Rev.* **30**, 66 (1927); *J. Am. Chem. Soc.* **52**, 3796 (1930).

¹⁰ See accompanying articles, *Phys. Rev.* **43**, 38–50 (1933).

of the light is recorded thus (?); while a full and unmistakable minimum is recorded by a plus sign (+).

After a given isotope has "come in" it con-

tinues to show at its particular scale reading throughout all subsequent increases in concentration. Eventually a concentration is reached beyond which no new minima occur, and the

TABLE I. *PbCl₂ from Joachimstahl: Observational record. Concentration: Grams of salt in 10¹² cc of water.*

Atomic mass	3.0	3.1	3.2	3.3	3.4	3.5	3.6	3.7	3.8	3.9	4.0	4.1	4.2	4.3	4.4	4.5	4.6	4.7	4.8	4.9	5.0	5.1	5.2	Magneto-optic scale reading
216	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	+	+	+	+	+	36.10
215	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	+	+	+	+	.19
214	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	+	+	+	+	.28
213	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	+	+	.40
212	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	+	+	+	+	.50
211	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	.59
210	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	.68
209	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	.78
208	?	+	+	+	+	?	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	.90
207	-	-	-	-	?	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	37.08
206	-	-	-	?	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	.30
205	-	-	-	-	-	-	?	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	.40
204	-	-	-	-	-	-	-	?	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	.50
203	-	-	-	-	-	-	-	-	?	+	+	+	+	+	+	+	+	+	+	+	+	+	+	.68
202	-	-	-	-	-	-	-	-	-	-	?	+	+	+	+	+	+	+	+	+	+	+	+	.79
201	-	-	-	-	-	-	-	?	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	.90

TABLE II. *Observational record PbCl₂ from Belgian Congo. Concentration: Grams of salt in 10¹² cc of water.*

Atomic mass	3.7	3.8	3.9	4.0	4.1	4.2	4.3	4.4	4.5	4.6	4.7	4.8	4.9	5.0	5.1	5.2	5.3	5.4	5.5	5.6	5.7	5.8	5.9	6.0	6.1	6.2	3.3	6.4	6.5	6.6	6.7	6.8	6.9	7.0	7.1	7.2	7.3	7.4	7.5	7.6	7.7	7.8	7.9	Magneto-optic scale reading			
216	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	36.10		
215	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	.19	
214	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	+	+	+	+	+	+	+	+	+	+	+	+	+	+	.28		
213	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	.40			
212	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	.50		
211	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	.59		
210	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	.68		
209	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	.78	
208	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	.90	
207	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	37.08	
206	?	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	.30
205	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	.40	
204	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	.50
203	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	.68
202	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	.79
201	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	?	.90

TABLE III. *PbCl₂ from Great Bear Lake: Observational record. Concentration: Grams of salt in 10¹² cc of water.*

Atomic mass	3.8	3.9	4.0	4.1	4.2	4.3	4.4	4.5	4.6	4.7	4.8	4.9	5.0	5.1	5.2	Magneto-optic scale reading
216	-	-	-	-	-	-	?	?	+	+	+	+	+	+	+	36.10
215	-	-	-	-	-	-	?	+	+	+	+	+	+	+	+	.19
214	-	-	-	-	-	-	-	-	-	-	-	-	?	+	+	.28
213	-	-	-	-	-	-	-	-	-	-	-	-	-	?	+	.40
212	-	-	-	-	-	-	-	-	-	-	-	-	-	?	+	.50
211	-	-	-	-	-	-	-	-	?	+	+	+	+	+	+	.59
210	-	-	-	-	-	-	-	-	-	-	-	-	-	?	+	.68
209	-	-	-	-	-	-	-	-	-	-	-	-	-	?	+	.78
208	-	-	-	?	?	+	+	+	+	+	+	+	+	+	+	.90
207	-	-	-	?	+	+	+	+	+	+	+	+	+	+	+	37.08
206	-	?	+	+	+	+	+	+	+	+	+	+	+	+	+	.30
205	-	-	-	-	-	-	-	?	+	+	+	+	+	+	+	.40
204	-	-	-	-	-	-	-	-	?	+	+	+	+	+	+	.50
203	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	.68
202	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	.79
201	-	-	-	-	-	-	-	-	?	+	+	+	+	+	+	.90

TABLE IV. *PbCl₂ from Kolm: Observational record. Concentration: Grams of salt in 10¹² cc of water.*

Atomic mass	9.3	9.4	9.5	9.6	9.7	9.8	9.9	10.0	10.1	10.2	10.3	10.4	10.5	10.6	Magneto-optic scale reading
216	—	—	—	—	—	—	—	?	+	+	+	+	+	+	36.10
215	—	—	—	—	—	—	—	?	+	+	+	+	+	+	.19
214	—	—	—	—	—	—	—	—	?	+	+	+	+	+	.28
213	—	—	—	—	—	—	—	—	—	?	+	+	+	+	.40
212	—	—	—	—	—	—	—	—	?	+	+	+	+	+	.50
211	—	—	—	—	—	—	—	+	+	+	+	+	+	+	.59
210	—	—	—	—	—	—	—	—	?	+	+	+	+	+	.68
209	—	—	—	—	—	—	—	+	+	+	+	+	+	+	.78
208	—	—	—	—	—	+	+	+	+	+	+	+	+	+	.90
207	—	—	—	—	+	+	+	+	+	+	+	+	+	+	37.08
206	—	—	?	+	+	+	+	+	+	+	+	+	+	+	.30
205	—	—	—	—	—	—	—	—	—	+	+	+	+	+	.40
204	—	—	—	—	—	?	+	+	+	+	+	+	+	+	.50
203	—	—	—	—	—	?	?	+	+	+	+	+	+	+	.68
202	—	—	—	—	—	—	—	—	+	+	+	+	+	+	.79
201	—	—	—	—	—	—	—	?	+	+	+	+	+	+	.90

number of minima are then interpreted as representing the number of isotopes of the particular element being studied, and it is further interpreted that the isotope existing in greatest abundance makes its appearance first. This is further substantiated by the fact that this technique is in agreement, in the case of lead, with that of Aston's mass-spectrograph, in demonstrating that the order of abundance in ordinary lead is 208, 206, 207, whereas in radioderived lead from a uranium mineral it is 206, 207, 208. As yet there has not been worked out a definite quantitative, numerical, relation between the position of a threshold appearance of an isotope in the table or on the graph and the relative concentration of that isotope within its group. Therefore, although it is known that the most abundant isotope makes its appearance first, we are, as yet, unable to establish the exact numerical relations existing among the members of an isotope group. Because of this, the general configuration of the graphs is more significant than the distances between the ends of the columns, but these distances undoubtedly reflect the orders of abundance of the respective isotopes.

In the case of uranium, the minimum which appeared at a scale setting of 29.62, and at the lowest concentration (2.1×10^{-12}), was interpreted as being caused by an isotope of mass 238 since this isotope is undoubtedly the most abundant one.¹¹ Because the scale reading 29.54 is in the direction of a heavier atom, that minimum was interpreted as being caused by a

¹¹ See also Aston, *Nature* **128**, 725 (1931).

uranium isotope possessing a mass greater than 238. By analogous reasoning the minimum at 29.65 is assigned to an isotope of uranium having a mass less than 238, and so on.¹² These isotopes of the respective elements are arranged according to their radioactive relationships in Fig. 1. The detailed reasons for this particular arrangement are given by Bishop¹³ elsewhere. Suffice it to say here that this represents what appeals to us as the most logical interpretation of all the existing data. The vertical columns are arranged from atomic number 92 to atomic number 81, each column containing all the isotopes of a particular atomic number, as determined by the Allison magneto-optic method. Each column is furthermore designated by the symbol of the element associated with that atomic number in the periodic table. Arrows pointing horizontally and to the right are used, in the interpretation here proposed, to indicate the loss of an alpha-particle while those pointing diagonally downward and toward the left indicate a beta-particle change.

The following discussion is derived from an inspection of Figs. 1-3.

URANIUM

There are eight atomic entities of atomic number 92, i.e., possessing 92 electrons in the planetary swarm. Four of these eight isotopes of ura-

¹² This elementary explanation is given thus in extenso because conversations have shown considerable confusion in the minds of many people concerning the manner of interpretation of these data.

¹³ Edna R. Bishop, *Radioactive Families*, Phys. Rev. **43**, 38 (1933).

ATOMIC No.	92	91	90	89	88	87	86	85	84	83	82	81
ELEMENT, TYPE	U	Pb	Th	Ac	Ra	VI	Ne	As	Po	Bi	Pb	Tl
DOMINANT FAMILY												
THORIUM	240		236		232						216	216
	236		232		228		224			212	212	208
										212	208	208
										208	204	204
ACTINIUM	239		235		231		227		223	219	215	215
	235		231		227		223		219	215	211	211
							219		215	211	207	207
							215		211	207	203	203
URANIUM	238		234		230		226		222	218	214	214
	234		230		226		222		218	214	210	210
									210	206	206	206
									206	202	202	202
UNNAMED	237		233		229		225		221	217	213	213
	233		229		225		221		217	213	209	209
							217		213	209	205	205
							213		209	205	201	201

FIG. 1. Suggested arrangement of the radioactive isotopes in four series.

nium appear to be primary and the remaining four secondary in the sense that the latter may be derived from the former by the loss of one alpha and two beta-particles, thus bringing them back to the same atomic number but with the loss of four units of mass. These primary uraniums together with their corresponding secondary isotopes are thought to constitute the head members of four families, or radioactive series. These are: the thorium series, actinium series, uranium series, and a fourth series as yet unnamed, whose head members are primary and secondary uranium isotopes of masses 237 and 233 respectively.

The most abundant first order uranium is U^{238} , which produces the most abundant first order thorium, Th^{234} (see respective graphs); this in turn produces the most abundant second order uranium, U^{234} , and also the second order thorium, Th^{230} . But all of Th^{234} passes through Th^{230} , some going by way of U^{234} and the remainder by way of Ra^{230} . These two thoriums occur at about the same order of abundance despite the short period of Th^{234} (24 days) and the long life of Th^{230}

(10^5 years). This may be accounted for by the fact that Th^{234} splits up in two ways and that there is interposed between it and Th^{230} the long-lived U^{234} (10^6 years). This series produces leads 214, 210, 206, and 202.

The next uranium in order of abundance is U^{239} , which is designated as the parent of the actinium series. This produces the first order thorium, Th^{235} , which is shown on the graph to be the next (in abundance) of the first order thoriums. This thorium, by the same scheme of interpretation, produces protoactinium Pa^{235} , which splits two ways, some going to actinium 231 and thence to Th^{231} while the remainder goes through U^{235} to the same Th^{231} . Just as U^{239} is next in order of abundance to primary U^{238} so secondary U^{235} is next after secondary U^{234} . But Th^{231} is nothing like as abundant, with respect to its primary Th^{235} , as Th^{230} is to its primary Th^{234} . This may

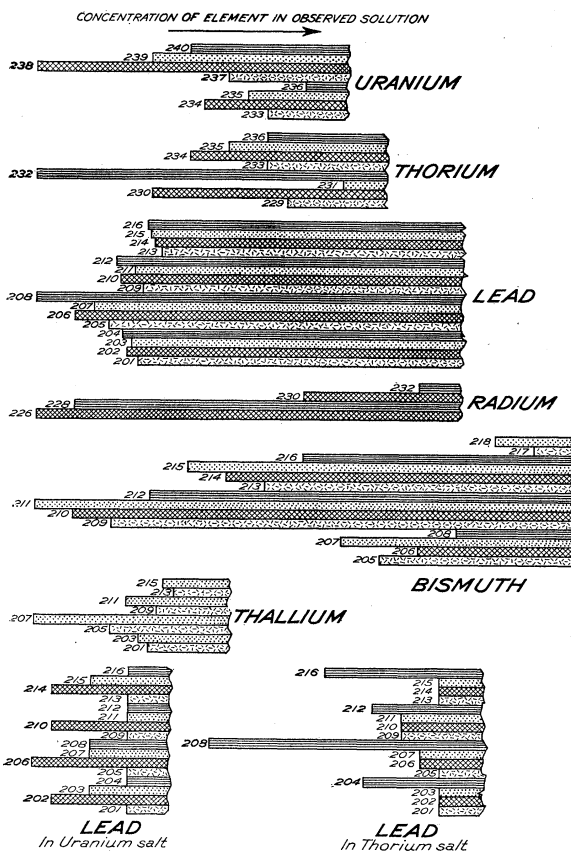


FIG. 2. Graphs of the number and order of abundance of the isotopes of uranium, thorium, lead, radium, bismuth and thallium in the observed solutions.

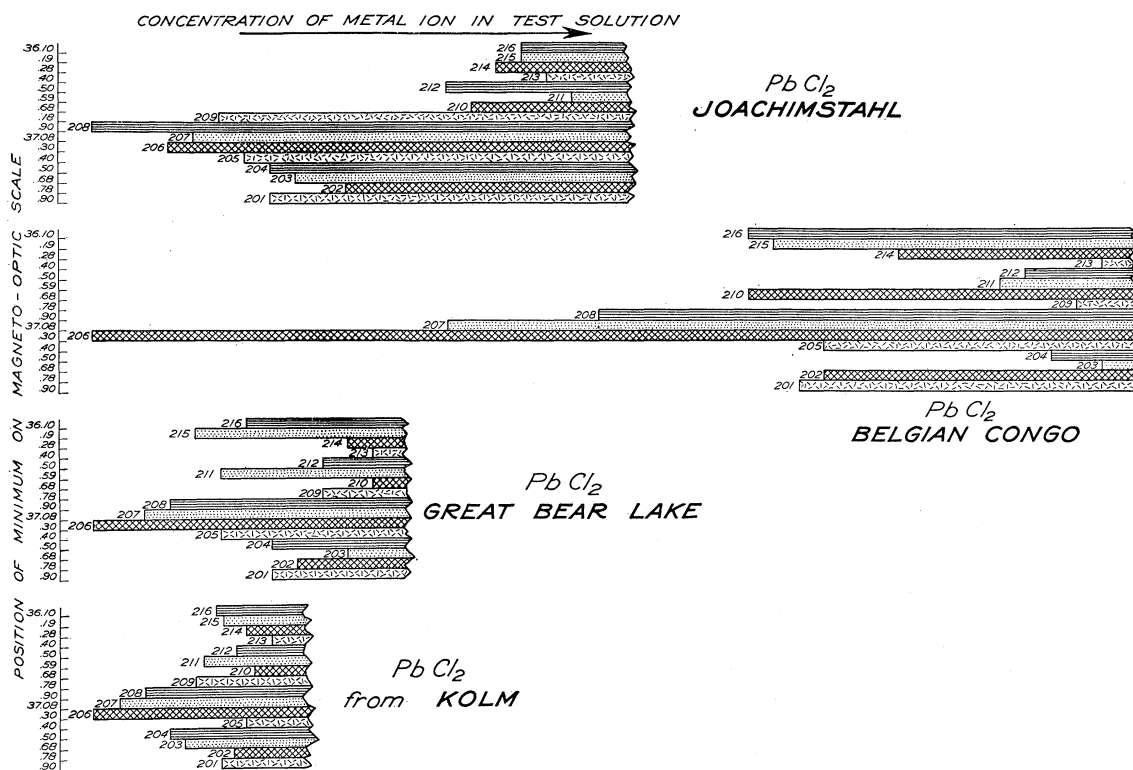


FIG. 3. Graphs of the order of abundance of the isotopes of certain radioactive leads.

be explained by the fact that all the material existing in Th^{235} does not pass through Th^{231} , for some goes by way of Ac^{231} ultimately to Pb^{215} , while only the remaining portion produces Th^{231} . This relationship is strikingly brought out by the graphs (Figs. 2 and 3) which, as has been stated, indicate the order of abundance of the respective isotopes. The much greater abundance of U^{238} over U^{235} gives Th^{234} a lead over Th^{235} despite the fact that Th^{235} breaks up with only one beta-particle, whereas Th^{234} gives off both alpha and beta-particles, and such substances are short-lived. Furthermore, the abundance of Th^{231} is decreased by virtue of its being a second beta-rayer, as all such substances are short-lived. This series produces the leads 215, 211, 207, and 203.

The next most abundant primary uranium is U^{240} , which is assumed to be the parent substance of the thorium series. This conception regards thorium as derived from an isotope of uranium. This is an old idea which was subsequently abandoned because no constant ratio of thorium to uranium seemed to exist. The explanation now may be that U^{240} probably has a life period which

is relatively shorter than U^{238} and quite possibly shorter than all the others. It produces Th^{236} which has a half life of 3.6 days, and this in turn goes to Ra^{232} of half life 6.7 years and U^{236} of half life 1.9 years. All of these periods are very short, but the material represented by these substances reassembles at Th^{232} which has the longest life period of all the known radioactive substances, i.e., 1.6×10^{10} years. These circumstances gave Th^{232} an independent status, for whereas early in its history it was associated with the rapidly disintegrating U^{240} (which is now mostly used up), it accumulated to such an extent that subsequent chemical changes, leachings and transportations, gave it the dominant position as the head of a family, in some cases appearing almost entirely dissociated from the uranium clan. This accounts for its irregular ratio in minerals with respect to the long-lived uranium as represented, for example, by U^{238} ($T = 4.5 \times 10^9$ years). This series produces leads 216, 212, 208, and 204.

The fourth and least abundant of all the series is the one headed by U^{237} which is the least abundant of all the primary uraniums. This is inter-

puted to produce the least abundant primary thorium Th^{233} which proceeds to U^{233} which is of low abundance. This may proceed to Th^{229} which bears the same relation to Th^{233} that Th^{231} does to Th^{235} , i.e., only part of the material of Th^{233} passes through Th^{229} ; and one would expect the ratio of abundance of Th^{229} to Th^{233} to be about the same as that of Th^{231} to Th^{235} . But it is not, and this is a discrepancy as yet unexplained. This series produces leads 213, 209, 205, and 201.

From the above it is evident that the relation of uranium to the lead derived from it is much more complex than was originally supposed. Because the U^{238} is at present by far the most abundant of the uranium isotopes it is the one that has been most studied, and this series has been very thoroughly worked out by orthodox methods. However, there are some uncertainties which these methods have failed to clear up, and it may be that in some cases they are due to the fact that the procedure used gave the experimenter a multiplicity of isotopes when it was thought that only one atomic species was involved. This possibility is briefly discussed by Bishop in the accompanying article. For example, the alpha-particles of different ranges which have been observed coming from the C'' members may not have come from one kind of nucleus but from several kinds.

LEAD

It is most significant that the order of abundance of the principal isotopes of ordinary and of radioderived lead is revealed by this method to be the same as that found by Aston with the mass-spectrograph. That is to say, the order in ordinary lead is 208, 206, 207, while in radioderived lead it is 206, 207, 208.

These facts and the configurations of the graphs are sufficient to demonstrate conclusively that ordinary lead has a different isotopic composition from that of radioderived lead, and that radioderived leads differ somewhat among themselves. These different isotopic compositions result from differences in age and also in the ratios existing among the isotopes of the parent element at the time of the formation of the mineral from which they were extracted, as will be developed later.

In his letter to *Nature* in August, 1927¹ Aston states: "There are indications that many other isotopes may be present in small proportions." He had fairly definite evidence of isotopes 209 and 205 and he suggested the possibility of 204 and 203. In a subsequent communication to *Nature* Aston reports eight isotopes of lead and states that he believes others exist.¹⁴ The magneto-optic technique reveals eight isotopes of masses progressively heavier than 208 and five of masses progressively lighter than 206; and it is a matter of the utmost significance that all the leads so far examined, both ordinary and radio-derived, have possessed all sixteen isotopes, i.e., they consist of the same isotopes, though these exist in different proportions in different samples.

The fact that both ordinary lead and radio-derived lead possess the same *number* of isotopes, of the same respective masses, is strong evidence that all the lead now on the earth was derived from the same ultimate source. If ordinary lead atoms came into being by whatever processes all other ordinary atoms did, and if this process is entirely distinct from and independent of radioactive processes as we know them, it seems extraordinary that exactly the same number of nuclear configurations should be produced by the two different and independent processes.

According to one school of thought the conditions existing in interstellar space presumably are favorable to the assembly of the respective fundamental units into various configurations and proportions to produce a variety of atomic nuclei. Some of these become very complex and when the accumulation and condensation of matter bring about the conditions obtaining in a star these more complex nuclei begin to break up into simpler configurations. Apparently this process had gone on to such an extent in our own sun that at the time the earth was formed from it there was already a considerable accumulation of lead atoms formed from the original uranium. These became mixed with the other atoms of which the earth is composed and have led an independent existence ever since. The remaining uranium atoms did likewise and both groups have since experienced the many vicissitudes of the geologic history of the rocks. According to this conception

¹⁴ Aston, *Nature* 129, 649 (1932).

the ordinary lead which we find today is in that isotopic condition to which it could attain by the further disintegration of the heavier isotopes into the lighter end-products during that period which has elapsed since the consolidation of this planet. Presumably this further disintegration has proceeded very slowly and by "soft" radiations, because the $\text{Pb}^{208, 204; 207, 203; 206, 202; \text{and } 205, 201}$ pairs of isotopes, though preponderating somewhat in ordinary leads, do not do so to a very great extent; and furthermore because no hard or easily detectable radiation has been noticed coming from ordinary lead.

Of the heavy lead isotopes, Pb^{214} (which presumably is Ra *B*) is best known, and this has been assigned a half life of 26.8 minutes. Such a short life would preclude its existence in ordinary lead today, and here our findings seemed incompatible with established evidence. However, an examination for uranium of a concentrated solution of the lead samples always revealed the presence of this element, which presumably was supplying the $\text{Pb}^{216, 215, 214, \text{ and } 213}$ which was observed.

An effort was made to prepare a lead salt which would be entirely free from this contaminating uranium, and though the treatment decreased the uranium content we have not up to the time of writing been able to eliminate the uranium entirely from a lead sample. A sample of radium *D*, kindly supplied by Mr. L. R. Hafstad of the Department of Terrestrial Magnetism, revealed leads 210, 206, and 202 in the order named, but no 214 whatever.

According to the above interpretation a young lead would be expected to be relatively weak in the lighter isotopes, i.e., those of mass 208 and less, whereas an old lead would possess these light isotopes to a somewhat preponderating degree. This condition would be best illustrated when the original parent uranium of the respective lead samples possessed the same isotopic concentrations at the beginning of the life of the particular minerals they are now in. This is unlikely. The uranium itself might have been young or old at the time it became fixed in the respective mineral. If young, it would have relatively more of isotope 240 and consequently the accumulated lead would now be richer in 208 than a lead produced from a uranium which had lost most of its

240 component before it became fixed in the mineral where we find it today. Such a uranium, being richer in 238 to begin with, would produce a lead relatively richer in the 206 isotope.

The unexpected results of this investigation make it clear that the isotopic relationships existing between uranium and the lead associated with it in minerals is much more complex than was previously supposed. It is apparent that there are at least three fundamental factors controlling the isotopic condition of any radioderived lead we may examine today and we do not have sufficient definite information about any of them. They are: (1) The isotopic condition of the uranium at the time of the formation of the mineral. (2) The rate of change of the many atoms involved between the uranium and its corresponding lead—and consequently the particular series as a whole. (3) The time that has elapsed since the formation of the mineral. There are, of course, other factors such as contamination, leaching, and so forth, of which we have little knowledge. Fortunately the $\text{U}^{238}\text{-Pb}^{206}$ series has been very thoroughly studied, and the rate of change of this series quite definitely established. Furthermore, Aston's experiments indicate that the U^{238} isotope comprises about 96 percent of the uranium existing today, and this isotope has probably predominated for a long time, so that the age determinations now being made are probably quite reliable. And those determinations which may be made, by using these two isotopes only, should be very close to the true value. We have hopes of soon devising some means of accurately determining the numerical relations existing among a group of isotopes. When this is done the ratio between U^{238} and Pb^{206} only need be considered.

It is now clear that other factors besides those of contamination and thorium content influence the chemically determined atomic weight of a given sample of lead, and it is apparent that a lead of atomic weight lower than 206 may exist.

Fig. 3 demonstrates that the lead from the Belgian Congo is exceptional, so far as our experience goes. Repeated rechecks of this material have failed to alter the configuration of this graph. Yet the graph for the uranium from which this lead came is not exceptional in any way.

The extraordinary preponderance of the three principal isotopes 206, 207, and 208 is noticeable, as is also the unusual amount of Pb^{206} with respect to the other two.

In this respect the graph agrees with Aston's measurements on a lead from this region in which he found the 206 isotope to be 93.3 percent of the whole, which is much greater than in any others he has reported. He found the 208 isotope to exist only to 0.02 percent, which is much less than one would expect from an inspection of the above graph. This is a striking example of the strange fact which Allison and his associates have consistently observed with respect to the isotopes of a given element, namely, that they "come in" too soon. If the graph actually pictures a relationship between the orders of abundance of Pb^{206} and Pb^{208} of the order of magnitude of the ratio of 93.3 to 0.02, then the remaining lead isotopes must exist in extremely small proportions of the whole. This, of course, is the conclusion already arrived at from many calculations of the atomic weight.

What particular conditions produced this sort of isotopic concentration are not obvious. If we assume that all lead is of radioderived origin and recall that the different series produce their respective leads at different rates, the conclusion is tenable that the ordinary lead now in existence has been in existence longer and has gone on farther toward an ultimate end condition, toward which the radioderived lead of our present day is now proceeding; and that the present isotopic condition of this ordinary lead is determined by the fact that somewhere back in its history it became separated from all or nearly all of its source of supply. In view of these considerations it seems fair to conclude that the radioderived lead of today is in an isotopic condition controlled by the isotopic condition of the uranium at the time of the formation of the containing mineral, the rate of production of the lead of the respective series, and the age of that particular mineral. Under such conditions the most rapidly produced isotope would gain in amount over its fellows until its source of supply was diminished or exhausted, whereupon the others would catch

up and possibly pass it, and the whole would ultimately attain to a condition which was controlled by the factors described above. This means that an inspection of the relative abundances of the respective isotopes of a certain lead affords no indication of its actual age; for, early in the history of events, there would exist only small differentials among the respective isotopes, like a group of runners at the beginning of a race, and very late in the process the differentials would again be small by virtue of the exhaustion of the source of supply of the more rapidly formed isotopes. Intermediate between these extremes there would be a time at which the most rapidly produced isotope would have its greatest accumulation with respect to its fellows. Possibly the Belgian Congo lead represents some such situation with respect to the other radioderived leads examined. The available literature gives their ages as: Kolm,¹⁵ 458 million years; Belgian Congo (Katanga)¹⁶ 600 million years; Great Bear Lake¹⁷ 1200 million years.

The graph of the Joachimstahl lead shows that this is contaminated by ordinary lead. This is revealed by the preponderance of the 208 isotope and the order of abundance of the principal isotopes, i.e., 208-206-207. This is in agreement with the fact that this deposit contains various ordinary lead minerals. This question of contamination is an important one in age determinations and it would seem that an isotopic examination is a reliable way of detecting its existence, though we do not yet know how small a contamination can be revealed by this method.

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¹⁵ R. C. Wells, *J. Wash. Acad. Sci.* **21**, 409 (1931).

¹⁶ *Nat. Res. Council Bull.* **80**, *Age of the Earth*, p. 388.

¹⁷ Private communication from Dr. J. P. Marble. (Preliminary calculation.)