Reports

Oceanic Basalt Leads: A New Interpretation and an Independent Age for the Earth

Abstract. *A modified form of a model proposed by Gerling and Shukolyukov is used to study the development of lead isotopic ratios in oceanic basalts. This modified model yields an age for the earth which is calculated to be 4530 ± 40 million years. The significance of this calculation is that it is independent of the knowledge of the ages of the samples studied. It is also found that although the source of the basalts is not generally homogeneous, the differentiation of this source from a closed system has occurred fairly recently geologically. The oldest time for the differentiation, 1230 ± 350 million years ago, has been found for Mid-Atlantic tholeiites.*

Recently reported lead isotope ratios and uranium, thorium, and lead concentrations in oceanic basalts suggest that the source of these basalts is not homogeneous. Tatsumoto (*1*) has stated that the results from Hawaii "strongly suggest that lead isotopic compositions in the magmas are different at their sources and that perhaps the upper mantle is not homogeneous, even under such a small island." He further indicates that the Hawaiian results contradict the conclusions of Stanton and Russell (*2*) and Russell and Farquhar (*3*) that conformable ore lead must come from an isotopically homogeneous subcrustal source.

The question of the homogeneity and "closed-system" nature of the upper mantle is also intimately related to the age of the earth. The accepted figure for this age, 4550 million years, has been recently disputed by Tilton and Steiger (*4*).

I wish to examine these topics here by making use of a graphical construction modified from that originally proposed by Gerling and Shukolyukov (*5*). This construction, particularly in the form presented here, provides an excellent graphical description of the relationship of the common and radiogenic lead contained in a U/Pb system.

Let Pb^{206} be the number of atoms of lead-206 and let U^{238} be the number of atoms of uranium-238 present in a rock of age t_1. Then, if $Pb^{206}{}_c$ is the number of common lead atoms (that is, the lead atoms which were not generated by the uranium-238 present in the rock) contained in the rock:

$$Pb^{206} = U^{238}(e^{\lambda t_1} - 1) + Pb^{206}{}_c \quad (1)$$

There is a similar equation for Pb^{207} present in the rock

$$Pb^{207} = U^{235}(e^{\lambda^1 t_1} - 1) + Pb^{207}{}_c$$

where λ and λ^1 are the decay constants for U^{238} and U^{235} respectively.

Dividing Eq. 1 by Pb^{204}, which is a nonradiogenic isotope, we get

$$\frac{Pb^{206}}{Pb^{204}} = \frac{U^{238}}{Pb^{204}}(e^{\lambda t_1} - 1) + \left(\frac{Pb^{206}}{Pb^{204}}\right)_c \quad (2)$$

It is well known that some Pb^{206} and Pb^{207} existed when the earth was formed. It is assumed that these lead isotope ratios are equal to those observed in the troilite phase of iron meteorites (see *6*):

$$\left(\frac{Pb^{206}}{Pb^{204}}\right)_0 = a_0 = 9.56$$

and

$$\left(\frac{Pb^{207}}{Pb^{204}}\right)_0 = b_0 = 10.42$$

Let us subtract a_0 from both sides of Eq. 2 and write

$$\frac{Pb^{206}}{Pb^{204}} - a_0 = \frac{Pb^{206*}}{Pb^{204}}$$

and

$$\left(\frac{Pb^{206}}{Pb^{204}}\right)_c - a_0 = \left(\frac{Pb^{206}}{Pb^{204}}\right)_{co}$$

Equation 2 may now be written in the form

$$\frac{Pb^{206*}}{Pb^{204}} = \frac{U^{238}}{Pb^{204}}(e^{\lambda t_1} - 1) + \left(\frac{Pb^{206}}{Pb^{204}}\right)_{co} \quad (3)$$

Following a similar procedure for Pb^{207}

$$\frac{Pb^{207*}}{Pb^{204}} = \frac{U^{235}}{Pb^{204}}(e^{\lambda^1 t_1} - 1) + \left(\frac{Pb^{207}}{Pb^{204}}\right)_{co} \quad (4)$$

We now combine Eqs. 3 and 4:

$$\frac{Pb^{206*}}{U^{238}} = (e^{\lambda t_1} - 1) + \frac{1}{\alpha}\left(\frac{Pb^{206}}{Pb^{207}}\right)_{co}\left\{\frac{Pb^{207*}}{U^{235}} - (e^{\lambda^1 t_1} - 1)\right\} \quad (5)$$

where

$$\frac{Pb^{206*}}{U^{238}} = \left(\frac{Pb^{206}}{Pb^{204}} - a_0\right)\frac{Pb^{204}}{U^{238}} \quad (6)$$

$$\frac{Pb^{207*}}{U^{235}} = \left(\frac{Pb^{207}}{Pb^{204}} - b_0\right)\frac{Pb^{204}}{U^{235}} \quad (7)$$

and

$$\alpha = \frac{U^{238}}{U^{235}} \text{ at present} = 137.8$$

In other words, a plot of

$$\frac{Pb^{206*}}{U^{238}} \text{ against } \frac{Pb^{207*}}{U^{235}}$$

for a number of rocks which were formed at the same time t_1, at which time varying amounts of common lead were incorporated into each rock, is a straight line of slope

$$\frac{1}{\alpha}\left(\frac{Pb^{206}}{Pb^{207}}\right)_{co}$$

I have assumed that the U^{238}/Pb^{204} ratio observed in the rock is representative of its source, which was formed t_1 years ago, and that the Pb^{206}/Pb^{207} ratio for common lead in each rock is the same.

Let us now superimpose a "concordia" curve represented by the equations

$$\frac{Pb^{206}}{U^{238}} = e^{\lambda t} - 1$$

and

$$\frac{Pb^{207}}{U^{235}} = e^{\lambda^1 t} - 1$$

on the graphical representation of Eq. 5. The straight line of Eq. 5 will intersect the concordia at two points. The physical significance of these intercepts may be established in the following way. The development of the common lead isotope ratios

$$\left(\frac{Pb^{206}}{Pb^{204}}\right)_c \text{ and } \left(\frac{Pb^{207}}{Pb^{204}}\right)_c$$

in a closed system from time t_0, the age of the earth, to a time t_1 may be represented by

$$\left(\frac{Pb^{206}}{Pb^{204}}\right)_c = a_0 + \mu(e^{\lambda t_0} - e^{\lambda t_1}) \quad (8)$$

and

$$\left(\frac{Pb^{207}}{Pb^{204}}\right)_c = b_0 + \frac{\mu}{\alpha}(e^{\lambda_1 t_0} - e^{\lambda_1 t_1}) \quad (9)$$

where $\mu = U^{238}/Pb^{204}$ in the closed system at present.

It follows from Eqs. 8 and 9 that

$$\left(\frac{Pb^{206}}{Pb^{207}}\right)_{co} = \frac{\alpha\,(e^{\lambda_1 t_0} - e^{\lambda_1 t_1})}{(e^{\lambda_2 t_0} - e^{\lambda_2 t_1})}$$

Hence

$$\frac{1}{\alpha}\left(\frac{Pb^{206}}{Pb^{207}}\right)_{co} = \frac{(e^{\lambda_1 t_0} - 1) - (e^{\lambda_1 t_1} - 1)}{(e^{\lambda_1 t_0} - 1) - (e^{\lambda_1 t_1} - 1)} \quad (10)$$

Since Eq. 10 represents the slope of the straight line which intersects the concordia, it is seen that if the lower intersection point occurs at t_1, then the upper intercept occurs at t_0. It may be seen from Eq. 5 that if no common lead has been added to the rock

$$\frac{Pb^{206*}}{U^{238}} = e^{\lambda_1 t_1} - 1$$

which is a point on the concordia at t_1. In other words, the lower intersection point of Eq. 5 with the concordia gives the age of formation of the uranium-thorium-lead system which is the immediate source of the leads analyzed. The upper intercept which occurs at a time t_0 is the age of the earth. The data used are summarized in Table 1.

Gerling and Shukolyukov (*5*), in their original model, did not correct for primordial lead. They interpreted the upper intercept as an upper limit to the time of the separation of uranium and lead in the earth's crust. Although this is a correct interpretation for the suites of young samples studied by Gerling and Shukolyukov (*5*), it should be pointed out that in their model the upper and lower intercepts can actually become interchanged in the case of very old samples.

Let us first of all test the proposed model by considering the case of the series of whole rocks from the Llano Uplift in Texas which were formed 1120 million years ago. At that time common lead, which had developed in a closed single-stage system from 4550 million years ago was added in varying amounts. The line in Fig. 1 is the least-squares fit (*7*) to the whole-rock data. The scatter of points about the line could be due either to a violation of closed system conditions in the whole rocks or to a multistage history for the common-lead component. In any case, the agreement between the intercept times and the ages obtained in the original interpretation is very good. The age of the earth calculated for this suite is 4530 million years. (Table 2).

Let us now apply our model to several other suites of rocks, the uranium and lead analyses for which have been published recently by Tatsumoto (*1, 8*). The uranium and lead analyses of the following suites were available to me: Hawaii (Fig. 2), Easter Island (Fig. 3), and Japan (Fig. 4). I have included the sample from the island of Oki Dogo, analyzed by Tatsumoto (*1*). There is ample evidence (*9*), however, that basalts from the Japan Sea side are markedly different from those of the Pacific side. The Pb/U ratios in Fig. 4 have been scaled by a factor of two so that their relationship to the concordia may be seen.

Tholeiites from the East Pacific rise and the Mid-Atlantic rise are shown in Fig. 5. Separate least-squares lines are shown, since I do not feel that it is justifiable to consider these tholeiites as belonging to one suite.

For Guadelupe Island, see Fig. 6. Consider first of all the information from Figs. 1 to 6 (and Table 2) which pertains to the age of the earth. The only assumption which I have made in plotting these figures is that the primordial lead abundances of the earth were coincident with those of iron meteorites. Providing then, that the common lead present in the rocks did develop in a closed system (Eqs. 8 and

Table 1. Uranium-to-lead atomic ratios and the isotopic composition of lead in samples from Llano, Texas; Mid-Atlantic Ridge; East Pacific Rise; Japan, and Hawaiian, Easter, and Guadelupe Islands. Data from Tatsumoto (*1, 8*) and Zartman (*16*).

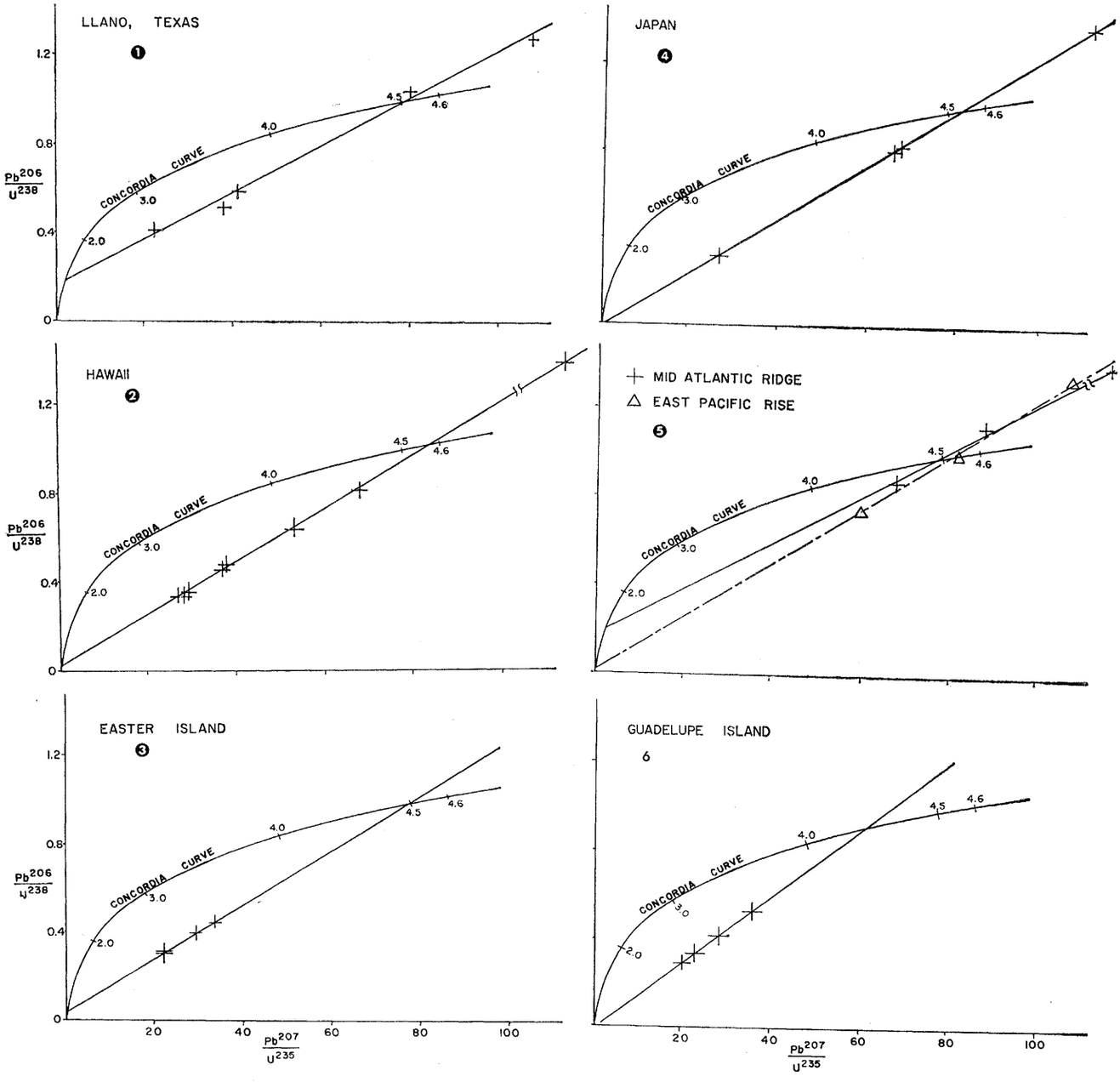
Sample No.*	Atomic ratios			Isotopic composition of lead (atomic ratio)	
	Observed	Modified†			
	U^{238}/Pb^{204}	Pb^{206}/U^{238}	Pb^{207}/U^{235}	Pb^{206}/Pb^{204}	Pb^{207}/Pb^{204}
		Llano Uplift, Texas			
3 gr	8.63	1.041	80.99	18.55	15.49
142 qdg	6.43	1.031	109.00	17.93	15.51
1 gn	33.67	0.407	22.38	23.27	15.89
21 gn	19.44	.526	37.63	19.78	15.73
53 gn	17.11	.595	41.71	19.75	15.60
		Mid-Atlantic Ridge			
AD2	7.9	1.128	89.31	18.47	15.54
AD3	5.6	1.476	126.00	17.82	15.54
AD5	10.5	0.882	69.03	18.82	15.68
		East Pacific Rise			
PD1	6.4	1.349	110.20	18.19	15.54
PD3	11.6	0.748	60.70	18.24	15.53
PD4	8.5	1.052	83.66	18.50	15.58
		Hawaiian Islands			
HM_c 1	5.2	1.640	136.7	18.09	15.58
HM_c 2	25.0	0.344	28.55	18.17	15.61
HM_c 3	10.5	.811	67.85	18.08	15.59
MH_c 4	19.7	.452	36.38	18.47	15.62
HM_c 5	13.2	.633	52.72	17.94	15.50
HM_c 6	27.2	.329	26.14	18.52	15.58
HM_c 7	26.0	.334	27.35	18.24	15.58
HM_c 8	19.2	.466	27.24	18.50	15.61
		Japan			
JCP 1	3.3	2.721	220.1	18.54	15.69
JHK 2	5.4	1.642	133.2	18.45	15.65
JHK 3	5.5	1.607	131.3	18.40	15.66
JHK 4	13.0	0.664	55.2	18.19	15.63
		Easter Island			
Pv 650	31.4	0.310	23.00	19.31	15.66
Pv 652	30.9	.314	23.01	19.25	15.58
Pv 653	21.1	.462	34.68	19.30	15.73
Pv 651	23.9	.407	30.27	19.28	15.67
		Guadelupe Island			
Gn 77	21.0	0.518	34.91	20.44	15.74
Gn 52	35.6	.301	20.56	20.79	15.73
Gn 22	33.1	.321	22.22	20.17	15.76
Gn 44	26.5	.401	27.30	20.18	15.67

* Sample No. is the number actually used by Zartman (*16*) in the case of the Llano uplift and by Tatsumoto (*1, 8*) for the remaining samples. † The modified ratios have been calculated with Eqs. 6 and 7.

9) and was incorporated into the rock at the time of the formation, the upper age is the age of the earth. This assumption is not unreasonable, considering the close fit of the points to straight lines and the fair agreement between the ages of rock formation calculated from the radiogenic and common-lead contributions (with the exception of Guadelupe).

The values for the age of the earth which I have calculated range from 4460 million years to 4580 million years (Table 2), with an average value of 4530 ± 40 million years. This is in excellent agreement with the age 4550 ± 70 million years obtained by Murthy and Patterson (*6*), and the age 4530 ± 30 million years obtained by Ostic, Russell, and Reynolds (*10*). The significance of the present calculation lies in the fact that no independent ages need be assigned to the samples studied. The only other age of the earth calculation which did not require independent age determinations is that by Ostic, Russell, and Reynolds (*10*), who used a number of galena samples which they assumed could be represented by a single growth curve. Their estimate is based, therefore, on an analysis of terrestrial common lead. The present calculation utilizes the information from both the common and radiogenic lead components contained in a Pb/U system and is correspondingly better determined. Common to all the methods is the definition of the age of the earth, which is the time at which terrestrial lead had the same isotopic composition as that in the troilite phase of iron meteorites.

In a recent article Tilton and Steiger (*4*) have calculated an age of the earth of 4750 million years. I believe that



Figs. 1–6. Modified Pb^{206}/U^{238} versus modified Pb^{207}/U^{235} ratios for: (1) the Llano Uplift, Texas; (2) Hawaiian Island basalts; (3) Easter Island basalts; (4) Japan basalts; (5) Mid-Atlantic Ridge and East Pacific Rise tholeiites; (6) Guadelupe Island volcanic rocks.

this estimate is not acceptable and I wish to discuss the reasons for their overestimation of this age (*11*).

Basically, Tilton and Steiger's method entails the assignment of an age value to a sample of galena of known isotopic composition and the use of Eqs. 8 and 9 to calculate a value of t_0. Inherent in this calculation is the assumption that the galena sample has developed in a closed single-stage system. The lead sample chosen by Tilton and Steiger is from Manitouwadge, Ontario. Very impressive evidence gathered from Rb/Sr, K/Ar, and U/Pb measurements exists in this area, which, as shown by Tilton and Steiger, indicates that the Manitouwadge ore is 2700 ± 100 million years old. Lead isotope ratios determined by various workers for galenas, feldspars and pegmatites from the area are shown graphically in Fig. 7. The line through the points representing the ratios in feldspar was interpreted by Tilton and Steiger as a "secondary isochron," conforming to the pattern expected from in situ uranium decay for 2700 million years. These authors did not, however, consider the implications of the lead isotope ratios reported by Ostic (*12*) and Slawson *et al.* (*13*) and shown in Fig. 7 for a galena sample from a vein near the Fox Creek fault in Geco property, Manitouwadge. [Tilton (*14*) does mention Ostic's (*12*) discussion of the possibility that Manitouwadge galena may contain a mixture of leads of two ages.] It is probable (*12*) that the Manitouwadge ore lead and the Geco vein lead are related in the sense that the vein lead migrated from one of the larger stratiform deposits, picking up radiogenic lead in the process, since there can be no in situ decay in the galena. The Pb^{207}/Pb^{206} ratio for a hypothetical in situ uranium decay from 2700 million years ago to the present is 0.186. The slope of the line in Fig. 7 joining the Manitouwadge and Geco leads is 0.189. In other words, the dispersion of lead isotope ratios illustrated in Fig. 7 is probably due to the combined effects of in situ uranium decay and a multistage history.

Slawson *et al.* (*13*) calculated a minimum value of 3100 million years for the source of the Manitouwadge lead. Ostic (*12*) estimated 2780 million years to be an upper limit to the age of deposition of the Manitouwadge ore lead. An age of 3100 million years is not unreasonable in this area. Catanzaro (*15*) has shown that zircons from southwestern Minnesota, although discordant, suggest a minimum crystallization age of 3300 million years.

Table 2. Various parameters obtained for the samples shown in Figs. 1 to 6 by least-squares analysis. Ages are millions of years.

Location of samples	Age of the earth	Intercept with Pb^{206}/U^{238} axis	Age from intercept	Slope $\frac{1}{\alpha}\left(Pb^{206}/Pb^{207}\right)_{co}$	Age from slope*
Mid-Atlantic Ridge	4510†	0.185 ± 0.067	1230	0.0103 ± 0.0007	1150
East Pacific Rise	4540†	0.021 ± 0.051	150	0.0121 ± 0.0006	200
Hawaii	4580†	0.013 ± 0.005	90	0.0119 ± 0.0001	320
Japan	4560†	−0.025 ± 0.007	−180	0.0125 ± 0.0001	0
Easter Island	4460†	0.017 ± 0.005	120	0.0128 ± 0.0003	−100
Llano Texas	4530†	0.153 ± 0.023	1028	0.0106 ± 0.0003	1000
Guadelupe Island	4270	−0.016 ± 0.007	−100	0.0153 ± 0.0003	−1800

* This age has been computed with Eq. 10 and the value $t_0 = 4530$ million years obtained in this work. † The mean of these ages is 4530 ± 40.

In summary, I believe that the lead isotope evidence implies that the lead in the Manitouwadge ore is remobilized lead which was differentiated from a closed system for at least 300 million years. Consequently, the use of closed-system Eqs. 8 and 9 with an age of 2700 million years is not valid in this case and results in a significant overestimation of the age of the earth.

I have chosen to consider the East Pacific rise tholeiites and the Mid-Atlantic ridge tholeiites separately since it is by no means clear that the source of these basalts is the same. Although each suite contains only three samples I feel that Fig. 5 illustrates that the sources of the Pacific and Atlantic tholeiites have had very different histories. In fact, whereas the source of the Pacific samples appears to have remained a closed system until recent times, the source of the Atlantic tholeiites was differentiated about 1230 ± 350 million years ago. In other words, the Mid-Atlantic ridge basalts have recorded a very old event even though they have been deposited in their present location in recent time.

The times of source differentiation for the Easter Island and Hawaiian basalts are 120 million years and 90 million years respectively. Until these times, according to the model, the common lead which was incorporated into these rocks developed in essentially closed single-stage systems.

I have thus far neglected the anomalous case of the Guadelupe Island

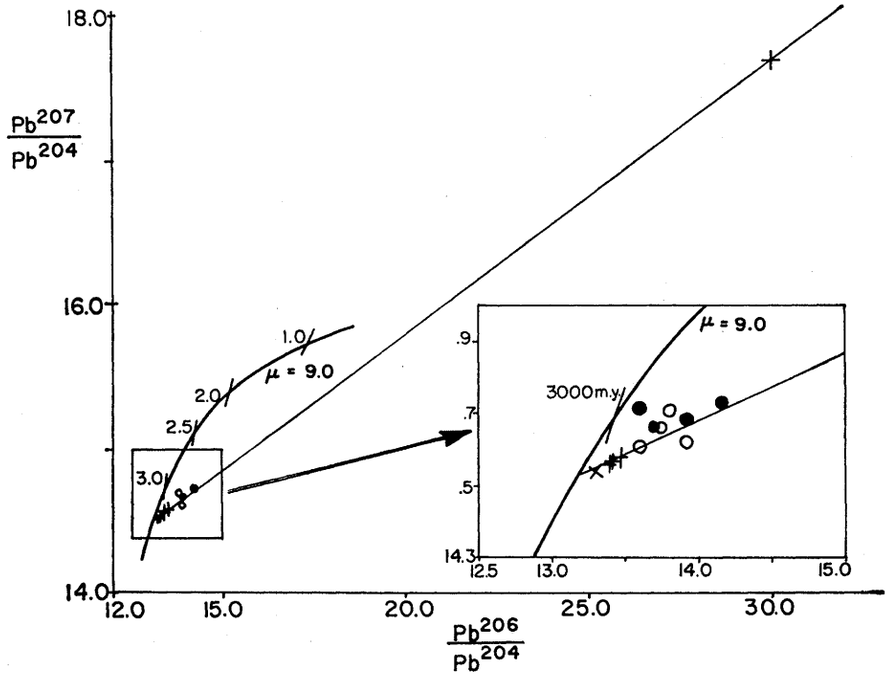


Fig. 7. Pb^{207}/Pb^{204} versus Pb^{206}/Pb^{204} for leads from Superior Province, North America. +, Manitouwadge galena (*12*); ×, Manitouwadge galena (*4*); ●, pegmatites near Manitouwadge (*4*); ○, granites and pegmatites from Rainy Lake and northern Minnesota (*17*).

illustrated in Fig. 6. The upper intersection in this figure gives an age of 4270 million years. The cases examined here, together with other evidence cited previously, strongly suggest an age of the earth of 4550 million years. Consequently, I conclude that the common lead emplaced into the Guadelupe rocks recently (due to the lower intercept in Fig. 6) was formed in at least a three-stage system as distinct from the essentially two-stage history of the common lead in the other suites.

The source of the basalt samples studied in this paper is the upper mantle. Superficially, the lead isotope and lead-uranium ratios for these samples tempt one to conclude that the upper mantle is heterogeneous with respect to lead-uranium and thorium on a global scale. Thus, Tatsumoto (*8*) has concluded that "the upper mantle is an open system chemically; thus, the lead in basalts extruded from the upper mantle could not have developed in a single closed system in contrast to the idea that conformable ore lead came from an isotopically homogeneous mantle." It is true that the immediate source rocks of many of these basalts are heterogeneous, but I believe that the model presented here shows that their association with such source rocks was relatively short lived (mostly less than 250 million years) and that they existed in closed systems for the greater part of the earth's history.

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Temperature-Dependence of the Polarity of Electrical Charges on Ice Crystals

Abstract. *The electrical polarity of ice crystals produced from a supercooled cloud is temperature-dependent. The charge polarity appears to be associated with the crystal habit. This phenomenon may be important in precipitation and cloud electrification processes.*

The relation between the crystal habit and the temperature of formation of ice crystals has been well documented (*1*, *2*). Workman and Reynolds (*3*) observed the electrical potential produced across an ice-water interface during the freezing of dilute aqueous solutions and suggested the relation of this phenomenon to cloud electrification. Experimental results indicate a relation between these phenomena. The electrical polarity and crystal habit of ice crystals produced from a fog of supercooled water droplets are related and are dependent on temperature and saturation ratio during crystal growth. In this note we offer the experimental results, and, without attempting to explain the fundamentals of the phenomenon, we suggest that it be considered as possibly having a bearing on thunderstorm electrification.

A fog of supercooled water droplets, with a typical diameter of less than 10 μ, was produced in a 24-m^3 chamber by the addition of approximately 300 g of steam. The resulting visibility was less than 2 m. After thermal equilibrium was attained, the supercooled fog was nucleated by the adiabatic expansion of a small quantity of moist air. This nucleation produced a number of ice crystals that were large enough to settle out within a few minutes. An excess of nucleation produced a relatively stable ice fog. The crystals were collected on a microscope slide, coated with formvar, and placed 0.5 mm below a grid of four parallel

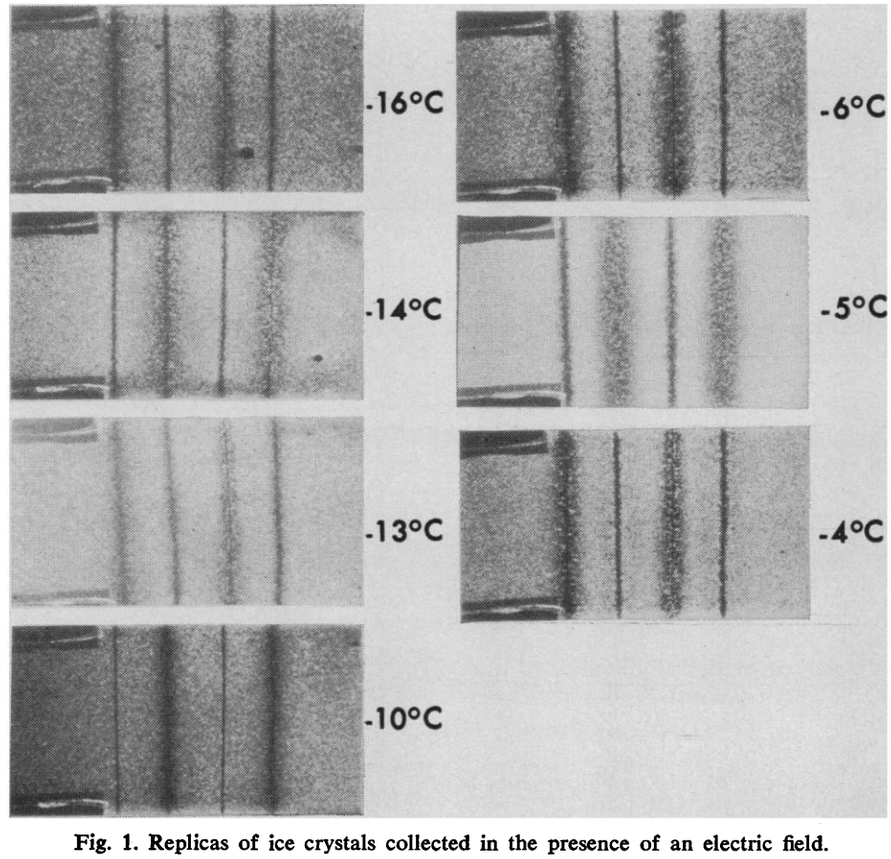

Fig. 1. Replicas of ice crystals collected in the presence of an electric field.