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THE SKEW FREQUENCY DISTRIBUTIONS AND THE FUNDAMENTAL LAW OF THE GEOCHEMICAL PROCESSES[1]

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ABSTRACT

The frequency distribution of concentrations of chemical elements has been investigated. Modern statistical methods for the analysis of large and small samples have been used. As a result of the work, the formulation of the fundamental law of the geochemical processes is proposed. This law can be applied in many cases to the investigation of deposits. The author points out the difference between "the distribution of the concentrations" in V. M. Goldschmidt's sense and "the probability distribution of the concentrations" of the present paper.

INTRODUCTION

One of the most valid problems of geochemistry is the investigation of the frequency distribution of the concentration of elements in igneous, metamorphic, and sedimentary rocks, minerals, soils, and organic matter. The positive solution of these problems will give the following: (*a*) a check of theoretical schemes of geochemistry by comparison of the theoretical probability distribution with empirical frequency distribution and (*b*) mean values of the concentrations of the elements and quantitative measurement of the correlation between the members of paragenesis estimations.

Richardson's paper (Richardson and Sneesby, 1922) is the first stage in the study of the frequency distribution in geochemistry, as it is known by the author. This paper won the broad approval of the petrologists and was theoretically interpreted by Richardson himself (1923). So in petrochemistry the theoretical sense of the frequency distributions was pointed out at the very beginning. Only five years before Richardson's paper was published, there had appeared A. A. Markov's mathematical proof of the physical sense of the probability distribution (Markov, 1917). Markov's very valuable paper aroused much interest in the mathematical literature, but these works remained completely unknown to geologists.

Since Richardson's paper was published, we have not had any frequency distribution investigation in geochemistry for seventeen years. But during these years F. J. Loevinson-Lessing published many papers on the frequency distributions of his "magmatic coefficients." These papers were stimulating to many Russian petrologists and to the author in particular.

In 1940 N. K. Rasumovsky wrote about the wide expansion of lognormal frequency distribution and particularly about the frequency distribution of the concentrations of elements. The wide expansion of lognormal distribution pointed out by Rasumovsky was taken into account by A. N. Kolmogorov, who gave his stochastic schemes by using modern methods (Kolmogorov, 1941). In 1948 Rasumovsky returned to his idea of lognormal distribution (Rasumovsky, 1948).

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During this period the lognormal distribution has been applied in the prospecting of gold.

Systematic analysis of frequency distributions for geochemical purposes investigated by the author have been carried out since 1943. In the course of time he studied the geochemical sense of the frequency distribution of the coefficients of porosity in Upper Permian beds of Buguruslan (Vistelius, 1945), the frequency distribution of six-valence sulfur in Paleozoic beds between the River Volga and the Ural Mountains (Vistelius, 1949). The stochastic foundation (Vistelius and Sarmanov, 1947) and statistical experimentation were presented in these papers as well as the application of the tests of the goodness of fit between the calculated functions and the observed frequencies. The basic conclusions of the author are as follows: the frequency distribution reflects the geochemical environment of sedimentation. The validity of the investigation of the frequency distributions was also pointed out by the author in some papers published later (Vistelius, 1948, 1952).

L. H. Ahrens' first paper (1953) on frequency distributions proposed the lognormal type of distribution and formulated his fundamental law of geochemistry. Three papers on this question were published by Ahrens later (1954*a*, 1954*b*, 1957). The whole scope of Ahrens' papers aroused the great interest of many geochemists in frequency distributions. His ideas gave rise to some critical articles (Aubrey, 1954, 1956; Chayes, 1954; Miller and Goldberg, 1955; Durovič, 1957). In Ahrens' papers the facts connected with frequency distributions were given for the first time as the material for the formulation of the law of geochemistry.

The importance of Ahrens' propositions, the disputability of them, and the many aspects of the problems and methods create a confused situation.

The author of this paper wished to avoid polemics and therefore preferred to analyze the frequency distributions but not to analyze general considerations given previously.

PRINCIPLES OF FREQUENCY ANALYSIS AND CHECK OF PREVIOUSLY PUBLISHED DATA

The frequency distribution analysis is a means of checking the hypothesis on the mechanism of the geochemical process which takes place for the chemical element x with the function of the probability distribution, $p(x)$. We formulate the problem as follows: The theory of the geochemical process concerning the element x gives the hypothesis H_0. This hypothesis is called the "null hypothesis H_0." We deduced from H_0 the probability distribution function of $p(x)$. Observations of the values of x as x_0, x_1, . . . , x_n give the frequencies $f(x)$. If the same function of difference,

$$\delta = F\left[f_i(x) - p_i(x) \right] , \qquad (1)$$

is small for the points i, we say that H_0 is not contradictory to our observations. If δ is large, we say that H_0 does not satisfy our observations and must be rejected as false.

In routine investigations we come across the following cases.

a) H_0 and consequently $p(x)$ are theoretically deduced from concrete investigations of the analyzed object, i.e., from the scheme of a geochemical process. In this case, in comparing $p(x)$ and $f(x)$, we can analyze the concrete sense of δ as well as that of $f(x)$. In other words, even increased values of δ can in some cases be explained and H_0 accepted.

b) H_0 and consequently $p(x)$ are not theoretically deduced from the geochemical sense of the process but are the explanation of frequencies $f(x)$ which are purely empirically smoothed by $p(x)$. In this case there are very severe restrictions on the value of δ. If these conditions are satisfied, we say that H_0 coincides with our observations (they coincide but are not proved!). If our conditions are not satisfied, H_0 is false.

In some cases we cannot say whether H_0 coincides or not with our observations; the values of δ are at the boundary of the zones of the rejection and acceptance of H_0. These conditions are in some way or other based on a number of observations.

INVESTIGATION OF THE PUBLISHED DATA

We shall study in detail the data published by N. K. Rasumovsky (1940) and L. H. Ahrens (1954*a*, *b*, 1957). The data of other authors (Aubrey, 1954; Durovič, 1957) have been published in the form of diagrams. The very small scale of these diagrams prevents us from analyzing them.

All the analyzed data may be divided into four groups.

The first group is given as the frequency distributions of Ga(g), Sc(d), K(g), Rb(g), Zn(g), V(d), La(g), Sc(g), Pb(g), V(g), and Cr(g).[2]

These distributions are given in the form of histograms by Ahrens (1954*a*, *b*, 1957). The cited histograms have only three classes with more than five observations in each class. Other classes have less than five observations in every class. According to statistics, we know that the goodness of fit of the calculated functions and the observed frequencies must be performed when every class has had more than five observations. Consequently, after excluding the classes with less than five observations, we obtain histograms with three classes only. But the normal distribution has three parameters; therefore, the number of degrees of freedom equals the quantity of classes minus three; as we have only three classes, we must have the highest goodness of fit with a calculated normal function with any three unequal numbers. So the first group of the data published cannot be examined.

The second group of the published observations contains histograms with frequencies having more observations than five in each class and with the number of classes more than three. Here are the data of Ahrens (1954*a*, *b*, 1957) on Cs(g), Mo(g), Ga(d), Co(d), Zr(d), Be(g), K(d), Rb(d), F(g), Mo in igneous rocks, Li in muscovite, and Rb in biotite, and of Rasumovsky (1940) on Cu, Pb, and Zn. These histograms have one or more degrees of freedom and can be investigated in some detail.

The smoothing of the observed frequencies in equal logarithmic values of classes by means of a normal curve for this distribution has been produced. After having calculated the ordinates of a normal curve, we compare them with the observed frequencies. For the sake of checking the goodness of fit of the calculated and observed data, we shall take Pearson's

$$\chi^2 = \sum_i \frac{[f(x_i) - p(x_i)]^2}{p(x_i)}, \qquad (2)$$

where χ^2 is the test of the goodness of fit, $f(x_i)$ is the observed and $p(x_i)$ the calculated frequencies by the lognormal H_0. As we know, the distribution of χ^2 with given degrees of freedom gives the level of confidence of H_0 in percentage. For the evaluation of the level of confidence, many tables were published which have at the head of the rows the degrees of freedom, whereas at the head of the columns are the levels of confidence, and within the table are the figures of χ^2. If we compare the calculated and observed frequencies without any theoretical background of H_0 (as it is in our case), we must take such great values of confidence as 50 per cent or more. Values of confidence smaller than 50 per cent give indefinite results of investigations; we can neither reject H_0 nor accept it.[3]

In table 1 observed and calculated frequencies, degrees of freedom, χ^2, and levels of confidence are given.

The technique of calculations of the nor-

[2] Cr(g) is chromium in granite; Sc(d) is scandium in diabase, etc.

[3] The value of the confidence level cannot be obtained in a pure theoretical way. As a rule, it must be revealed by experience. We have no data in our case about the mutual independence of observations. In such cases it is necessary to be very cautious in the acceptance of a new hypothesis. The above points out why we have accepted H_0 with the confidence level higher than 50 per cent and we suppose the 15 per cent level insufficient for acceptance. In our case the common confidence level used for independent observations and equal to 5 per cent cannot be used for acceptance of H_0.

mal curve by means of the observed $\bar{x}$ and S are illustrated in table 2. By repeating calculations similar to those of table 2, the reader can check all the other data in table 1.

The data from table 1 tell us that the lognormal H_0 can be more or less accepted for Cu in ores of Altai, Rb in biotite, Li in muscovite, K in diabase, Mo and Be in granites. The lognormal H_0 must be rejected for molybdenum in igneous rocks, Zr in diabase, and Pb in the ores of Altai. We must also reject the lognormal H_0 for As in granites and Cr in diabase. The geometrical peculiarities of these histograms (Ahrens, 1954, 1957) have no features of a normal curve, which is obvious without

TABLE 1

FREQUENCY DISTRIBUTIONS OF SOME CHEMICAL ELEMENTS (y IS OBSERVED AND y' IS CALCULATED FREQUENCIES) (LOGNORMAL H_0)

Elements Studied	Observed (y) and Calculated (y') Frequencies	Frequencies of the Classes										No. of Observations	χ^2	Degrees of Freedom (k)	Confidence Coefficients
		1	2	3	4	5	6	7	8	9	10				
(L. H. Ahrens)															
K in diabases	y	3	7	9	12	5	4					40	0.51	1	$0.50>P_k>0.30$
	y'	9.1		10.6	10.7	9.6									
Rb in diabases	y	2	7	3	7	8	4	3				34	2.81	1	$0.10>P_k>0.05$
	y'	6.2		14.3		6.7	6.8								
F in granites	y	1	7	15	20	7	3	1				54	1.73	1	$0.20>P_k>0.10$
	y'	8.8		14.7	16.2	14.3									
Mo in igneous rocks	y	1	5	6	35	24	8	1	1			81	9.03	2	$0.02>P_k>0.01$
	y'	4.3		14.3	26.2	23.5	12.7								
Ga in diabases	y	5	26	17	7	2						57	3.52	1	$0.10>P_k>0.05$
	y'	7.4	19.7	20.9	9.0										
Co in diabases	y	2	1	5	18	22	8	1				57	2.47	1	$0.20>P_k>0.10$
	y'	11.2			17.5	17.4	10.9								
Zr in diabases	y	2	6	7	25	9	3					52	7.43	1	$0.01>P_k>0.005$
	y'	6.5		13.9	17.4	14.2									
Rb in biotites	y	15	30	51	51	20	5					172	0.98	3	$0.90>P_k>0.80$
	y'	12.6	32.8	53.7	46.2	21.0	5.7								
Cs in granites	y	1	4	9	3	6	4	1				28	2.76	1	$0.10>P_k>0.05$
	y'	5.1		6.1	12.8		4.0								
Mo in granites	y	1	5	8	22	14	9	1	1			61	2.50	2	$0.30>P_k>0.20$
	y'	5.4		11.4	17.5	15.7	11.0								
Be in granites	y	1	4	6	16	9	6	5				47	2.44	2	$0.30>P_k>0.20$
	y'	4.5		8.1	12.1	11.6	10.7								
Li in muscovites	y	6	12	22	16	12	5	9				82	2.85	3	$0.50>P_k>0.30$
	y'	7.0	10.9	17.0	19.0	15.0	13.1								
(N.K. Rasumovsky)															
Zn	y	13	33	125	242	247	138	83	1			882	8.20	4	$0.10>P_k>0.05$
	y'	9.0	42.0	128.1	231.0	245.9	154.4	71.6							
Pb	y	1	9	36	100	111	148	178	123	91	19	816	35.20	6	$0.001>P_k$
	y'	12.9		32.4	76.2	133.7	173.6	168.0	120.3	64.2	34.7				
Cu	y	2	25	65	127	198	168	76	24	2	1	688	4.40	4	$0.50>P_k>0.30$
	y'	23.2		66.6	142.5	189.2	156.0	79.7	30.8						

TABLE 2

EXAMPLE OF CALCULATIONS OF NORMAL CURVE BY MEANS OF THE OBSERVED $\bar{x}$ AND S (EQUAL LOGARITHMIC CLASSES)

Logarithmic Classes	No. of Classes (x)	Frequencies Observed (y)	$x-\bar{x}$	$t=(x-\bar{x})/s$	$\phi(t_i)$	$\phi(t_i)-\phi(t_{i-1})$	y'	$\Delta=\|y-y'\|$	Δ^2	Δ_2/y'
			$-\infty$	$-\infty$	−0.5000					
0.01– 0.03.......	1	2				0.0056	3.85			
		} 27	−3.594	−2.535	−0.4944		} 23.18	3.82	14.59	0.75
0.03– 0.06.......	2	25				0.0281	19.33			
			−2.594	−1.829	−0.4663					
0.60– 0.125......	3	65				0.0968	66.60	1.60	2.56	0.04
			−1.594	−1.124	−0.3695					
0.125–0.250......	4	127				0.2071	142.48	15.48	239.63	1.68
			−0.594	−0.419	−0.1624					
0.250–0.500......	5	198				0.2750	189.21	8.79	77.26	0.41
			+0.406	+0.286	+0.1126					
0.500–1.000......	6	168				0.2268	156.04	11.96	143.04	0.92
			+1.406	+0.992	+0.3394					
1.000–2.000......	7	76				0.1158	79.67	3.67	13.47	0.17
			+2.406	+1.697	+0.4552					
2.000–4.000......	8	24				0.0366	25.18			
			+3.406	+2.402	+0.4918					
4.000–8.000......	9	2 } 27				0.0073	5.02 } 30.82	3.82	14.59	0.47
			+4.406	+3.107	+0.4991					
8.000–16.000.....	10	1				0.0009	0.62			
			$+\infty$	$+\infty$	+0.5000					
Total..........	....	688	....	....	....	1.0000	688.00	....	....	4.44

$\bar{x}\simeq 5.094$, $S=1.418$, $\chi^2=4.44$, $k=4$, $0.5>P_k(\chi^2)>0.3$

making any calculations. All the other distributions given by Ahrens (1954*a*, *b*) and the distribution of Zn given by Rasumovsky have such frequencies that they do not permit acceptance or rejection of the lognormal H_0. These data are quite indefinite (for example, the frequency distribution of Zn has a confidence coefficient between 5 and 10 per cent).

The third group of observations contains Ahrens' numerical data which are given in table 1 of this paper (Ahrens, 1954*a*). These data cannot be analyzed by a frequency grouping as Ahrens (1954*a*) did because the number of his observations is too small. But they can be analyzed by one of the newest methods, which we shall give later (Dunin-Barkowsky and Smirnov, 1955).

According to our H_0, we postulate that there exists a normal frequency distribution of $x = \log z$. By H_0 let

$$N[u(x); 0, 1] = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{u(x)} e^{-x^2/2} dx . \quad (3)$$

In this case and only in this case the function $u(x)$ is linear and is

$$u = \frac{x - a}{\sigma}, \quad (4)$$

where a is the mathematical expectation of x and σ is the standard deviation; in equation (4) we estimate a by $\bar{x}$ and σ by $\hat{s}$, where $\hat{s}$ is the unbiased statistic

$$\hat{s} = \sqrt{\frac{1}{n-1}\left[\Sigma x_i^2 - \left(\frac{\Sigma x_i}{n}\right)^2\right]} \quad (5)$$

and n is the number of observations. At our H_0 for logarithms z we have a straight line for the point $u(x)$ on the co-ordinate plane (x, u). Let us take our logarithms of concentrations of element in an ascending order and designate the points of the rise of the empirical function by $\tilde{u}_i$. It is obvious that the points $(x, \tilde{u}_i)$ near the straight line $u(x)$ are on the plane (x, u) for normal distribution.

The word "near" we understand thus:

The standard deviation of values $\tilde{u}_i$ of the observed distribution is normal and is given as

$$\sigma_{\tilde{u}_i} \approx \frac{1}{g(u_i)} \sqrt{\frac{0.25 - [\phi(u_i)]^2}{n}}, \quad (6)$$

where $g(u_i)$ is the probability density and $\phi(u_i)$ is the area of normal distribution. The tables of the normal probability distribution are to be found in every handbook of statistics. Using (6), we can find the function $\mathcal{E}(\sigma_{\tilde{u}_i})$, and by means of $\mathcal{E}(\sigma_{\tilde{u}_i})$ we shall establish the bounds of the confidence intervals for $\tilde{u}_i$ with the confidence coefficient, p. Taking the confidence coefficient p, we state that all the points of observation which lie between the straight line $u(x)$ and the curves $\mathcal{E}(\sigma_{\tilde{u}_i})$ are those "near the straight line $u(x)$"; all points which lie outside $\mathcal{E}(\sigma_{\tilde{u}_i})$ are "far from the straight line $u(x)$." If the points $\tilde{u}_i$ lie near $u(x)$, we accept H_0 with the confidence coefficient p. If the points $\tilde{u}_i$ lie outside $\mathcal{E}(\sigma_{\tilde{u}_i})$, we reject H_0 with the confidence coefficient $1 - p$. If the points $\tilde{u}_i$ lie near or on $\mathcal{E}(\sigma_{\tilde{u}_i})$, we can neither reject nor accept H_0.

In table 3 an example of calculations is given.

In table 4 the results of the calculations of $\tilde{u}_i$, u_i, and $\sigma\tilde{u}_i$ are given. In graphic form they are repeated in figure 1. From table 4 and figure 1 it was concluded that the lognormal H_0 for Ga must be rejected because some points of observations lie outside $\mathcal{E}(\sigma_{\tilde{u}_i})$. The H_0 can be accepted for Sc. Other distributions of the points (for Zr, Pb, La, V, Cr [?] in Canadian granites) are situated near 15 per cent confidence limits. For this reason we cannot reject the lognormal H_0; but the 15 per cent confidence coefficient is very small for the empirical function—that is why one cannot seriously accept the lognormal H_0. The situation is such that our confidence limits can be reached by many skew distributions and, in particular, by lognormals (Rasumovsky, 1940; Ahrens, 1954), cubic roots (Chayes, 1954), and so on.

The fourth and last group of published data is K/Rb in igneous rocks and chondrites; Rb_2O/Tl_2O in potassium minerals;

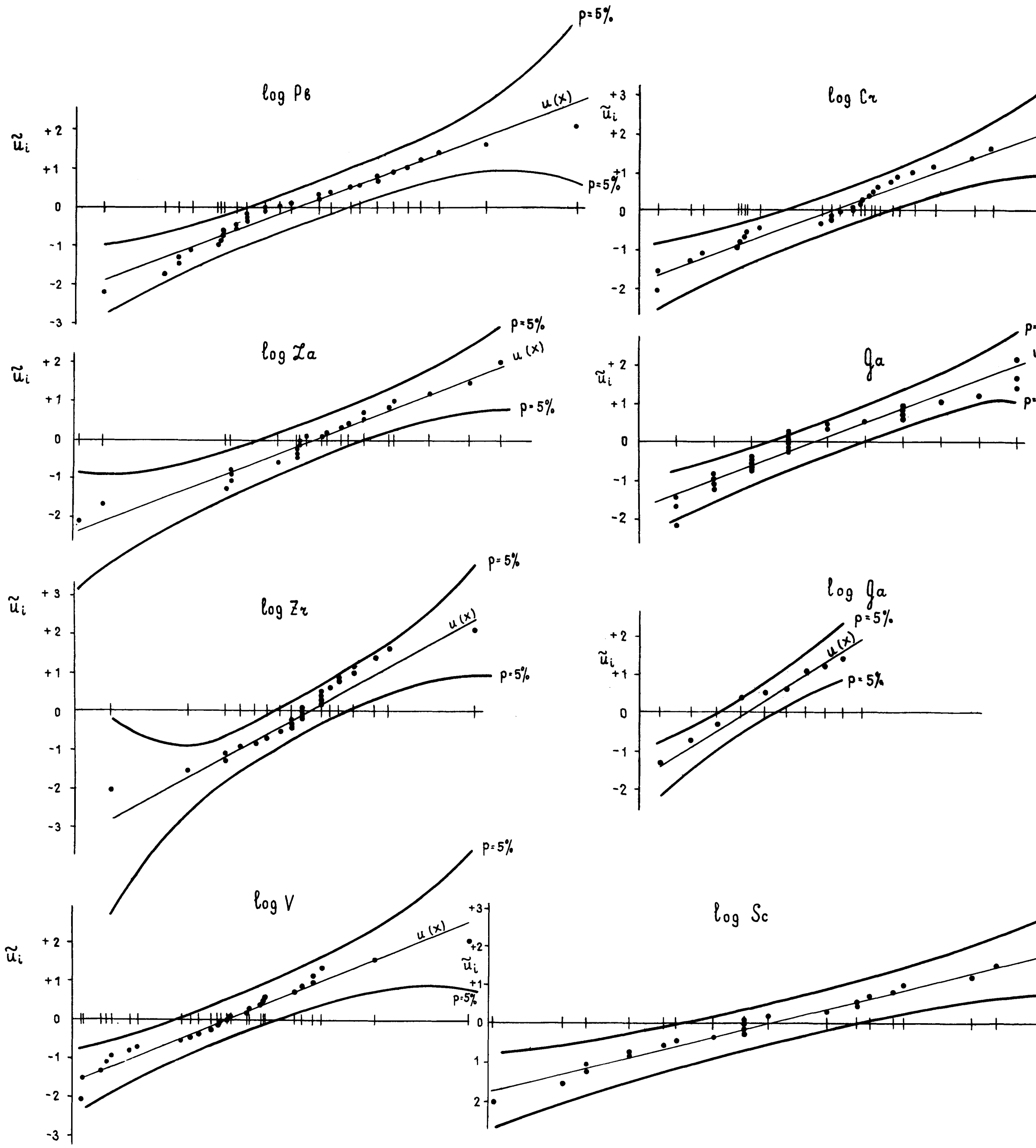

FIG. 1.—The straight-line diagrams for Ga, Cr, Sc, La, V, Pb, and Zn in Canadian granites. The dots are $\tilde{u}_i$; curv
$\mathcal{E}(\sigma_{\tilde{u}_i})$, are given for 5 per cent confidence limits (ρ) of $\tilde{u}_i$; they are plotted from the line $u(x)$ for the demonstrati

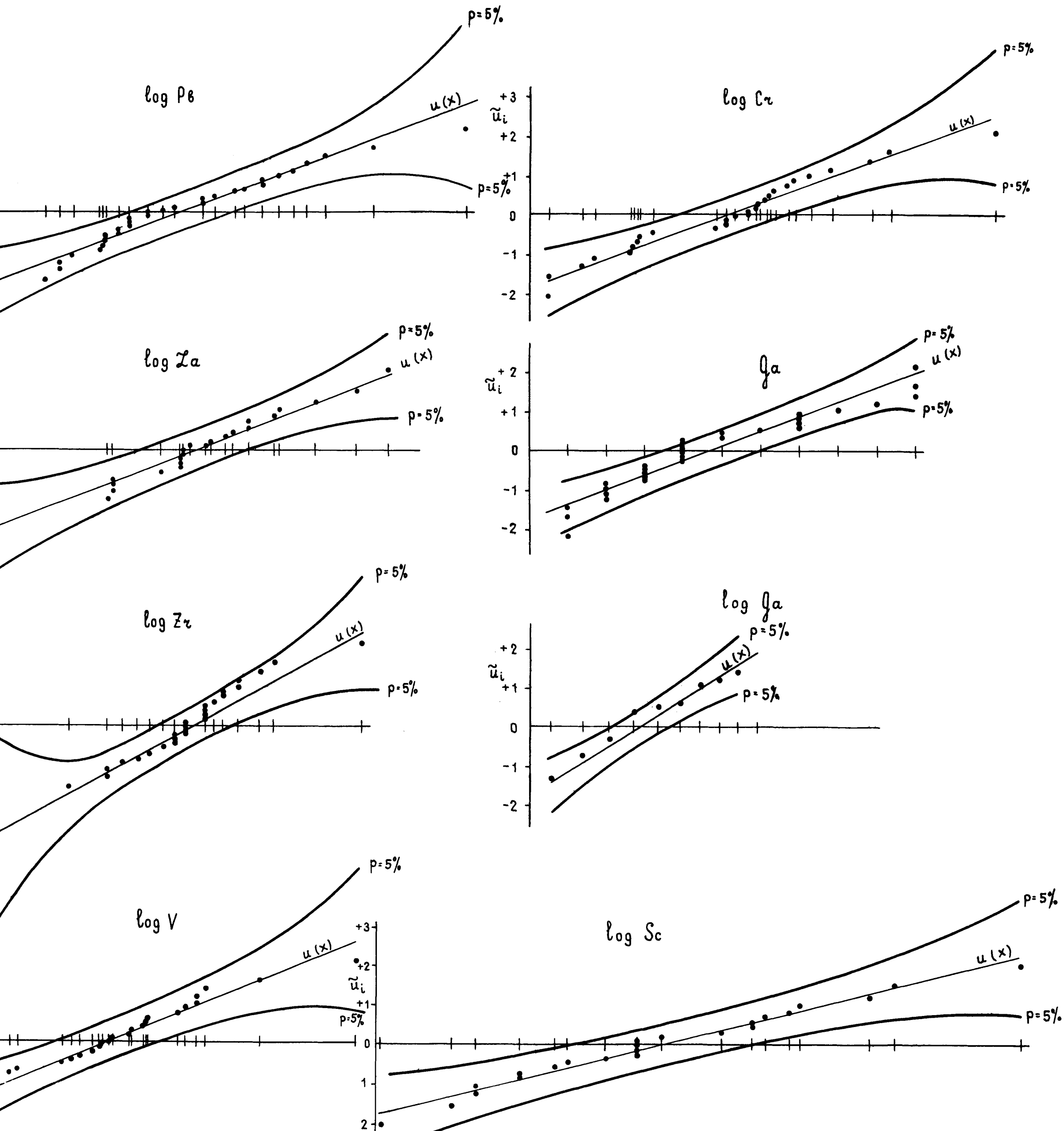

FIG. 1.—The straight-line diagrams for Ga, Cr, Sc, La, V, Pb, and Zn in Canadian granites. The dots are $\tilde{u}_i$; curves, $\sigma_{\tilde{u}_i}$), are given for 5 per cent confidence limits (ρ) of $\tilde{u}_i$; they are plotted from the line $u(x)$ for the demonstration.

TABLE 3

EXAMPLE OF CALCULATIONS u, $\tilde{u}$, AND $\sigma_{\tilde{u}}$

No.	Sc	ln 10Sc	$(100/n)(i-0.5)$	$\tilde{u}_i+5$	$\tilde{u}_i$	$X_i-\bar{X}$	$u_i=(X_i-\bar{X})/\hat{s}$	$\phi(u_i)$	$g(u_i)$	$\sqrt{0.25-[\phi(u_i)]^2}$	$\sqrt{n}\cdot g(u_i)$	$\|\tilde{u}_i-u_i\|$	$\sigma_{\tilde{u}_i}$	$\|\tilde{u}_i-u_i\|$ $1/\sigma_{\tilde{u}_i}$	$1.96\sigma_{\tilde{u}_i}$
1.....	1.3	2.5649	2.174	2.980	−2.020	−1.6576	−1.754	0.4603	0.0857	0.1952	0.4110	0.266	0.475	0.560	0.931
2.....	2.0	2.9957	6.522	3.488	−1.512	−1.2268	−1.298	0.4029	0.1718	0.2961	0.8239	0.214	0.359	0.596	0.704
3.....	2.3	3.1355	10.870	3.767	−1.233	−1.0870	−1.150	0.3749	0.2059	0.3308	0.9875	0.083	0.335	0.248	0.657
4.....	2.3	3.1355	15.218	3.973	−1.027	−1.0870	−1.150	0.3749	0.2059	0.3308	0.9875	0.123	0.335	0.367	0.657
5.....	3.0	3.4012	19.566	4.143	−0.857	−0.8213	−0.869	0.3076	0.2735	0.3942	1.3117	0.012	0.301	0.040	0.590
6.....	3.0	3.4012	23.913	4.290	−0.710	−0.8213	−0.869	0.3076	0.2735	0.3942	1.3117	0.159	0.301	0.528	5.900
7.....	3.7	3.6109	28.261	4.425	−0.575	−0.6116	−0.647	0.2412	0.3236	0.4379	1.5519	0.072	0.282	0.255	0.553
8.....	4.0	3.6889	32.609	4.549	−0.451	−0.5336	−0.565	0.2140	0.3401	0.4519	1.6311	0.114	0.277	0.411	0.543
9.....	5.0	3.9120	36.957	4.667	−0.333	−0.3105	−0.329	0.1289	0.3779	0.4831	1.8123	0.004	0.267	0.014	0.523
10.....	6.0	4.0943	41.305	4.780	−0.220	−0.1282	−0.136	0.0541	0.3953	0.4971	1.8958	0.084	0.262	0.321	0.514
11.....	6.0	4.0943	45.653	4.890	−0.110	−0.1282	−0.136	0.0541	0.3953	0.4971	1.8958	0.026	0.262	0.099	0.514
12.....	6.0	4.0943	50.000	5.000	0.000	−0.1282	−0.136	0.0541	0.3953	0.4971	1.8958	0.136	0.262	0.519	0.514
13.....	6.0	4.0943	54.349	5.110	+0.110	−0.1282	−0.136	0.0541	0.3953	0.4971	1.8958	0.246	0.262	0.939	0.514
14.....	7.0	4.2485	58.697	5.220	+0.220	+0.0260	+0.028	0.0112	0.3988	0.4999	1.9126	0.192	0.261	0.736	0.512
15.....	10.0	4.6052	63.045	5.333	+0.333	+0.3827	+0.405	0.1573	0.3675	0.4747	1.7625	0.072	0.269	0.268	0.527
16.....	12.0	4.7875	67.392	5.451	+0.451	+0.5650	+0.598	0.2251	0.3336	0.4464	1.5999	0.147	0.279	0.527	0.547
17.....	12.0	4.7875	71.740	5.575	+0.575	+0.5650	+0.598	0.2251	0.3336	0.4464	1.5999	0.023	0.279	0.082	0.547
18.....	13.0	4.8675	76.088	5.710	+0.710	+0.6450	+0.683	0.2527	0.3159	0.4314	1.5150	0.027	0.285	0.095	0.559
19.....	15.0	5.0106	80.436	5.857	+0.857	+0.7881	+0.834	0.2978	0.2818	0.4016	1.3515	0.023	0.297	0.077	0.582
20.....	16.0	5.0752	84.784	6.027	+1.027	+0.8527	+0.902	0.3165	0.2656	0.3870	1.2738	0.125	0.304	0.411	0.596
21.....	24.0	5.4806	89.132	6.233	+1.233	+1.2581	+1.331	0.4084	0.1645	0.2884	0.7889	0.098	0.366	0.268	0.717
22.....	28.0	5.6348	93.480	6.512	+1.512	+1.4123	+1.495	0.4325	0.1305	0.2508	0.6259	0.017	0.401	0.042	0.786
23.....	60.0	6.3969	97.828	7.020	+2.020	+2.1744	+2.301	0.4893	0.0283	0.1030	0.1357	0.281	0.759	0.370	1.684

$\bar{x}\approx 4.2225$; $\hat{s}\approx 0.9449$; $P(|x-\bar{x}|>0.939\ \sigma_{\tilde{u}})\approx 0.35$

TABLE 4

CHECK OF LOGNORMAL AND NORMAL (FOR GALLIUM) H_0

LOGNORMAL H_0

La				Ga				Sc				Pb			
log La	$\tilde{u}_i$	u_i	$\sigma_{\tilde{u}_i}$	ln Ga	$\tilde{u}_i$	u_i	$\sigma_{\tilde{u}_i}$	ln Sc	$\tilde{u}_i$	u_i	$\sigma_{\tilde{u}_i}$	ln Pb	$\tilde{u}_i$	u_i	$\sigma_{\tilde{u}_i}$
1.1139.....	−2.020	−2.307	0.765	2.5649...	−2.154	−1.497	0.340	2.5649...	−2.020	−1.754	0.475	3.6376...	−2.14	−1.837	0.435
1.1761.....	−1.512	−2.073	0.613	2.5649...	−1.675	−1.497	0.340	2.9957...	−1.512	−1.298	0.359	4.0073...	−1.66	−1.255	0.303
1.5051.....	−1.233	−0.833	0.297	2.5649...	−1.418	−1.497	0.340	3.1355...	−1.233	−1.150	0.335	4.0943...	−1.40	−1.118	0.285
1.5185.....	−1.027	−0.783	0.293	2.6391...	−1.230	−1.025	0.270	3.1355...	−1.027	−1.150	0.335	4.0943...	−1.21	−1.118	0.285
1.5185.....	−0.857	−0.783	0.293	2.6391...	−1.078	−1.025	0.270	3.4012...	−0.859	−0.869	0.301	4.1744...	−1.06	−0.992	0.270
1.5185.....	−0.710	−0.783	0.293	2.6391...	−0.946	−1.025	0.270	3.4012...	−0.710	−0.869	0.301	4.3175...	−0.93	−0.766	0.251
1.6435.....	−0.575	−0.312	0.272	2.6391...	−0.831	−1.025	0.270	3.6109...	−0.575	−0.647	0.282	4.3438...	−0.81	−0.725	0.248
1.6902.....	−0.451	−0.136	0.264	2.7081...	−0.724	−0.587	0.236	3.6889...	−0.451	−0.565	0.277	4.3820...	−0.70	−0.665	0.244
1.6902.....	−0.333	−0.136	0.264	2.7081...	−0.626	−0.587	0.236	3.9120...	−0.333	−0.329	0.266	4.3820...	−0.60	−0.665	0.244
1.6990.....	−0.220	−0.103	0.264	2.7081...	−0.533	−0.587	0.236	4.0943...	−0.220	−0.136	0.262	4.4427...	−0.51	−0.569	0.239
1.7160.....	−0.110	−0.039	0.264	2.7081...	−0.445	−0.587	0.236	4.0943...	−0.110	−0.136	0.262	4.4427...	−0.42	−0.569	0.239
1.7160.....	0.000	−0.039	0.264	2.7081...	−0.360	−0.587	0.236	4.0943...	0.000	−0.136	0.262	4.4998...	−0.33	−0.479	0.235
1.7559.....	+0.110	+0.112	0.263	2.7726...	−0.278	−0.178	0.223	4.0943...	+0.110	−0.136	0.262	4.4998...	−0.25	−0.479	0.235
1.7709.....	+0.220	+0.168	0.264	2.7726...	−0.197	−0.178	0.223	4.2485...	+0.220	+0.028	0.261	4.4998...	−0.16	−0.479	0.235
1.8062.....	+0.333	+0.301	0.267	2.7726...	−0.118	−0.178	0.223	4.6052...	+0.333	+0.405	0.269	4.6052...	−0.08	−0.313	0.229
1.8261.....	+0.451	+0.376	0.270	2.7726...	−0.039	−0.178	0.223	4.7875...	+0.451	+0.598	0.279	4.6052...	0.00	−0.313	0.229
1.8692.....	+0.575	+0.538	0.277	2.7726...	+0.039	−0.178	0.223	4.7875...	+0.575	+0.598	0.279	4.7005...	+0.08	−0.163	0.226
1.8692.....	+0.710	+0.538	0.277	2.7726...	+0.118	−0.178	0.223	4.8675...	+0.710	+0.683	0.285	4.7875...	+0.16	−0.026	0.225
1.9345.....	+0.857	+0.784	0.295	2.7726...	+0.197	−0.178	0.223	5.0106...	+0.857	+0.834	0.297	4.9416...	+0.25	+0.216	0.227
1.9494.....	+1.027	+0.841	0.300	2.7726...	+0.278	−0.178	0.223	5.0752...	+1.027	+0.902	0.304	4.9416...	+0.33	+0.216	0.227
2.0414.....	+1.233	+1.187	0.345	2.8332...	+0.360	+0.207	0.224	5.4806...	+1.233	+1.331	0.366	5.0106...	+0.42	+0.325	0.230
2.1461.....	+1.512	+1.582	0.425	2.8332...	+0.445	+0.207	0.224	5.6348...	+1.512	+1.495	0.401	5.1358...	+0.51	+0.522	0.237
2.2304.....	+2.020	+1.899	0.531	2.8904...	+0.533	+0.570	0.237	6.3969...	+2.020	+2.301	0.759	5.1930...	+0.60	+0.613	0.241
				2.9444...	+0.626	+0.913	0.259					5.2983...	+0.70	+0.778	0.252
				2.9444...	+0.724	+0.913	0.259					5.2983...	+0.81	+0.778	0.252
				2.9444...	+0.831	+0.913	0.259					5.3936...	+0.93	+0.928	0.264
				2.9444...	+0.946	+0.913	0.259					5.4806...	+1.06	+1.061	0.278
				2.9957...	+1.078	+1.239	0.296					5.5607...	+1.21	+1.192	0.294
				3.0445...	+1.230	+1.549	0.351					5.6699...	+1.40	+1.364	0.321
				3.0910...	+1.418	+1.844	0.431					5.9661...	+1.66	+1.830	0.433
				3.9010...	+1.675	+1.844	0.431					6.5221...	+2.14	+2.706	1.018
				3.0910...	+2.154	+1.844	0.431								

TABLE 4—*Continued*

Lognormal H_0												Normal H_0			
Cr				Zr				V				Ga			
log Cr	$\tilde{u}_i$	u_i	$\sigma_{\tilde{u}_i}$	log Zr	$\tilde{u}_i$	u_i	$\sigma_{\tilde{u}_i}$	log V	$\tilde{u}_i$	u_i	$\sigma_{\tilde{u}_i}$	Ga	$\tilde{u}_i$	u_i	$\sigma_{\tilde{u}_i}$
0.3010	−2.086	−1.697	0.422	1.6990	−2.086	−2.829	1.290	0.7404	−2.086	−1.542	0.381	13	−2.154	−1.356	0.314
0.3010	−1.593	−1.657	0.422	1.9031	−1.593	−1.673	0.415	0.7482	−1.593	−1.527	0.377	13	−1.675	−1.356	0.314
0.4771	−1.325	−1.377	0.346	2.0000	−1.325	−1.124	0.306	0.8451	−1.325	−1.334	0.338	13	−1.418	−1.356	0.314
0.5441	−1.128	−1.256	0.324	2.0000	−1.128	−1.124	0.306	0.8751	−1.128	−1.274	0.328	14	−1.230	−0.985	0.266
0.7243	−0.967	−0.929	0.283	2.0414	−0.967	−0.890	0.279	0.9031	−0.967	−1.218	0.319	14	−1.078	−0.985	0.266
0.7404	−0.828	−0.900	0.280	2.0792	−0.828	−0.676	0.262	1.0000	−0.828	−1.025	0.293	14	−0.946	−0.985	0.266
0.7634	−0.704	−0.858	0.276	2.1139	−0.704	−0.479	0.252	1.0414	−0.704	−0.943	0.285	14	−0.831	−0.985	0.266
0.7782	−0.590	−0.831	0.274	2.1461	−0.590	−0.297	0.245	1.2788	−0.590	−0.471	0.251	15	−0.724	−0.614	0.237
0.8451	−0.483	−0.710	0.265	2.1761	−0.483	−0.127	0.242	1.3222	−0.483	−0.384	0.248	15	−0.626	−0.614	0.237
1.1761	−0.381	−0.109	0.242	2.1761	−0.381	−0.127	0.242	1.3617	−0.381	−0.306	0.245	15	−0.533	−0.614	0.237
1.2304	−0.282	−0.011	0.241	2.1761	−0.282	−0.127	0.242	1.4314	−0.282	−0.167	0.242	15	−0.445	−0.614	0.237
1.2304	−0.187	−0.011	0.241	2.2041	−0.187	+0.033	0.241	1.4624	−0.187	−0.104	0.242	15	−0.360	−0.614	0.237
1.2788	−0.093	+0.077	0.241	2.2041	−0.093	+0.033	0.241	1.4771	−0.903	−0.076	0.241	16	−0.287	−0.243	0.224
1.3424	0.000	+0.193	0.243	2.2041	0.000	+0.933	0.241	1.5185	0.000	+0.007	0.241	16	−0.197	−0.243	0.224
1.3424	+0.093	+0.193	0.243	2.2041	+0.093	+0.033	0.241	1.5315	+0.093	+0.032	0.241	16	−0.118	−0.243	0.224
1.3802	+0.187	+0.261	0.244	2.2553	+0.187	+0.323	0.246	1.6232	+0.187	+0.215	0.243	16	−0.039	−0.243	0.224
1.3979	+0.282	+0.293	0.245	2.2553	+0.282	+0.323	0.246	1.6335	+0.282	+0.235	0.244	16	+0.039	−0.243	0.224
1.4314	+0.381	+0.354	0.247	2.2553	+0.381	+0.323	0.246	1.6990	+0.381	+0.388	0.247	16	+0.118	−0.243	0.224
1.4472	+0.483	+0.383	0.248	2.2553	+0.483	+0.323	0.246	1.7076	+0.483	+0.383	0.248	16	+0.197	−0.243	0.224
1.4771	+0.590	+0.437	0.250	2.2788	+0.590	+0.456	0.251	1.7160	+0.590	+0.400	0.248	16	+0.287	−0.243	0.224
1.5441	+0.704	+0.559	0.255	2.3010	+0.704	+0.582	0.257	1.8751	+0.704	+0.716	0.271	17	+0.360	+0.128	0.222
1.5798	+0.828	+0.623	0.259	2.3010	+0.828	+0.582	0.257	1.9138	+0.828	+0.793	0.271	17	+0.445	+0.128	0.222
1.6021	+0.967	+0.664	0.262	2.3424	+0.967	+0.816	0.273	1.9731	+0.967	+0.911	0.281	18	+0.533	+0.498	0.232
1.7709	+1.128	+0.970	0.287	2.3424	+1.128	+0.816	0.273	1.9731	+1.128	+0.911	0.281	19	+0.626	+0.869	0.255
1.9777	+1.325	+1.345	0.348	2.3979	+1.325	+1.131	0.307	2.1584	+1.325	+1.280	0.329	19	+0.724	+0.869	0.255
2.0792	+1.593	+1.530	0.378	2.4314	+1.593	+1.319	0.335	2.3010	+1.593	+1.564	0.386	19	+0.831	+0.869	0.255
2.6128	+2.086	+2.498	0.858	2.6128	+2.086	+2.346	0.732	2.7983	+2.086	+2.556	0.919	19	+0.946	+0.869	9.255
												20	+1.078	+1.240	0.296
												21	+1.230	+1.611	0.300
												22	+1.418	+1.981	0.480
												22	+1.675	+1.981	0.480
												22	+2.154	+1.981	0.480

Sr/Ca in limestones (Ahrens, 1954*b*); Ni/Fe, Zn/Fe, Cu/Fe, and Mn/Fe in pyrrhotites (Durovič, 1957). These frequency distributions are in contradiction to the lognormal H_0 for K, Rb, Sr, etc., because we know no reason why their ratio with other elements should be lognormal. It must be admitted that the problem of the probability distribution of products of random variables and their ratios is very complicated, especially for correlated random variables. It can be illustrated by the simplest products of two variables, each of them having a normal distribution and being correlated with one another (Aroian, 1947). In our case we have, according to H_0, lognormally distributed random variables (Ahrens, 1954*b*, 1957) with a strong correlation (K-Tl, for example) between them. In this case we do not know the distribution function of the ratio. In the case of Sr/Ca and Durovič's example (1957) we have the minor elements Sr, Ni, Zn, Cu, Mn, and the major ones, such as Ca in limestones and Fe in pyrrhotites. For the minor elements we can propose the lognormal H_0, but for the major elements this cannot be done from any general considerations, as was pointed out by Aubrey (1956). It is obvious that if there are great quantities of the element in any minerals or rocks—like Fe in pyrrhotites—the expectation of a great positive asymmetry is hardly real. New data on this question have not been published; therefore, we do not know what probability distributions will have ratios of minor to major elements of rocks or minerals.

Neither is it clear why $F(z)$ must be lognormal if $F(x)$ and $F(y)$ are lognormal and

$$F(z) = F(x) * F(y) .$$

where $*$ is the designation of the composition (Cramer, 1946), as was proposed by Rasumovsky for the distributions of Zn + Pb, every one of which has a lognormal distribution according to his hypothesis.

Our analysis of the published data is completed.

The lognormal distribution with a high confidence coefficient is a very rare case. The lognormal distribution can be rejected in very rare cases. In a great number of cases we can neither reject nor accept the lognormal H_0. The problem must be treated according to more sufficient data. This treatment will be given in the second part of this paper.

ANALYSIS OF SKEWED DISTRIBUTIONS

From the materials examined in the previous publications it follows that in many cases the frequency distributions of the concentrations of elements have positive skewness. These distributions can sometimes be sufficiently approximated by a lognormal function. Some of these distributions have a large skewness but are not lognormal. The fact is that the great positive skewness is widely observed and must be interpreted from the geochemical point of view. Two schemes can be given for the explanation of the skewness of distributions.

The first scheme has been cited by Miller and Goldberg (1955) taken from Cramer's handbook (1946). This scheme was proposed by Kaptein and Van Uven in the years 1903 and 1916. Its sense is the following: Let us have a concentration, X, which may be regarded as the sum of a large number of impulses $z_1, \ldots, z_n$. These impulses are mutually independent and act in the order of their subscripts. Let the increase produced by the impulse z_ν be assumed to be directly proportional to z_ν and to the concentration X. The concentration X_ν results from the action of $\nu - 1$ of the preceding impulses.

In this case

$$X_{\nu+1} - X_\nu = k\, z_\nu X_\nu ,$$

and hence

$$z_1 + \ldots + z_n = \frac{1}{k} \sum_{1}^{n} \frac{X_{\nu+1} - X_\nu}{X_\nu} . \quad (7)$$

If this process is discontinuous,

$$w = \frac{1}{k} \int_{X_1}^{X_2} \frac{dt}{g(t)} , \quad (8)$$

and w will be normally distributed, so that we shall have a lognormal distribution. It is obvious from equation (8) that we shall get a lognormal distribution only when $g(t)$ is a simple linear function, i.e., $g(t) = at$. In other cases we shall have various functions which would be very troublesome to use in normal distributions.

From the foregoing, we have only one population. This population is a lognormal population, and it occurs independently whether we have a whole batholith or a little bit of it.

The analyzed scheme assumes one frequency distribution function independently, whether we have sampled a piece of the rock or a geological body formed by this rock.

The second scheme is the following: The geochemical process at every separate moment gives a concentration of the elements in the conditions of the central-limit theorem (Cramer, 1946). In the course of time the mean values of the deposited concentrations increase. At the same time the values of variances also increase. The variability of the mean values and variances have a linear positive correlation as an approximate estimate of their relations. In other words, the probability density, $f(x)$, is

$$f(x) = \sum_i p_i f_i(x) , \qquad (9)$$

where $f_i(x)$ is normal density, p_i is the function of weight of summarized densities, and i is the stage of the geochemical process put in the order of time.[4]

In this case we have a skewed function $f(x)$, but this function, whether lognormal or not has only one certain feature—the positive skewness of $f(x)$ when p_i is given the order of non-increasing values.

The principal difference of the last scheme from lognormal is the following: the frequency distribution functions for one separate stage of the geochemical process[5] are normal. The frequency distribution functions for all joint products of the geochemical process have a positive skewness (we called these distributions "joint distributions").

The given schemes can be checked in an experimental way. For this purpose we must investigate the following questions: (*a*) whether the skewed distributions of the concentration of elements for the joint products of the geochemical process are lognormal and, if these distributions are not lognormal, whether we have a positive linear correlation between $\bar{x}$ and s for these distributions, and (*b*) whether the positively skewed and not lognormal distributions of joint products of the geochemical process have normal distributions for fixed stages of this geochemical process.

THE JOINT DISTRIBUTIONS, THEIR AVERAGES AND STANDARD DEVIATIONS

As an example of the joint distribution, we have investigated the distribution of phosphorus in granitic rocks. For this reason we compiled analytical data of P_2O_5 in the granites of the world which contain more than 60 per cent SiO_2. The frequency distribution for P_2O_5 in granitic rocks of Switzerland is given in figure 2. This distribution is valuable because the sampling was produced on a small territory and gives many analytical data. As we can see from figure 2, the analyzed distribution of P_2O_5 has a great positive skewness but is not a lognormal one, as is indicated by the estimations of its statistics (shown in fig. 2). So the first given distribution of the element cited by Ahrens (1957, p. 206) has a positive skewness but is not lognormal.

Table 5 gives the values of the averages of P_2O_5 and its standard deviations in some regions of the world. These figures are calculated from a number of analyses collected from many papers. We shall give the list of the papers used in our next article.

Proceeding as before (Vistelius, 1948, 1958), we got a coefficient of correlation be-

[4] The process of growing of the mean values and variances can be continuous.

[5] Let us name them "local distributions."

tween the average contents of P_2O_5 in samples and their standard deviation as 0.56. The equation of regression of the standard deviation on averages is as follows:

$$s = 0{,}601\,\overline{P_2O_5} + 0.0538\ .$$

From our analysis it is obvious that P_2O_5 has a positive skew distribution and a positive correlation between the average values and standard deviations of its distributions.

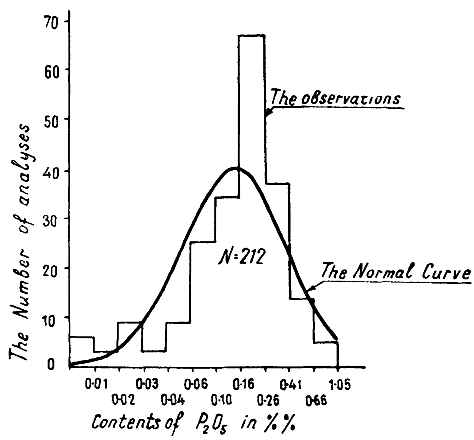

FIG. 2.—Frequency distribution of P_2O_5 in the granitic rocks of Switzerland (with $SiO_2 > 60$ per cent).

JOINT DISTRIBUTIONS VERSUS LOCAL DISTRIBUTIONS

In figure 3 there is given a histogram of the frequency distributions of Na_2O obtained by 4,788 analyses of igneous rocks (Washington, 1917). As shown in figure 3, the distribution of Na_2O in igneous rocks has great positive skewness and is not a lognormal one. So the joint distribution of Na_2O is a skew and not a lognormal one. In figure 4 is given the distribution of Na_2O in the sample of 200 analyses of the basalts of the world. This distribution is also skewed, but the skewness is smaller than in the previous case.

From these examples the following conclusions can be drawn. All the rocks of the world are a collection of results of a very wide range of stages of the geochemical process. Their joint distribution is very skew. All the basalts of the world are also a collection of the results of the range of stages of the geochemical process, but their range of the geochemical process is narrower than the range of those of all rocks. If we turned our attention to the skewness of the analyzed distributions, we could obtain, without any calculations, the notion that the skewness of the joint distribution of all the rocks is greater than that of the joint distribution of basalts.

We shall formulate the next hypothesis from observations in the following way: local distributions are normal, whereas the joint distributions are skewed. We checked our hypothesis by the following procedure.

In table 6 are given the figures for Na_2O concentrations in the basalts of the world. We placed in one row of table 6 the analyses with very similar contents of SiO_2. They are given for basalts taken from one definite place. In the columns of table 6 are given the analyses of basalts from the different geographical locations and with different compositions. We have rocks varying from leucite basalts to nepheline-melilite rocks and from taitites to andesite basalts.

For the understanding of the local distributions we proceed as follows (Dunin-Barkovsky and Smirnov, 1955).

Let us designate the percentage of Na_2O in basalts (table 6) by x and let

$$\tau_i = \frac{x_i - \bar{x}}{\hat{s}}, \qquad (10)$$

where $\bar{x}$ and $\hat{s}$ are designated as previously. In the values of τ we have a statistic which is free of the mean value of the distribution and its standard deviation but contains skewness and excess of distribution. We obtain values of τ by sliding from the upper left corner of table 6 to the lower right corner by four rows and then again to the left side of the table.

According to Cramer (1946), we know that

$$\phi(\tau) = \frac{\Gamma[(n-1)/2]}{\sqrt{\pi(n-1)}\,\Gamma[(n-2)/2]} \cdot \left(1 - \frac{\tau^2}{n-1}\right)^{(n-4)/2} \quad (11)$$

for $|\tau| < \sqrt{n-1}$.

But we have taken $n = 4$, so on the right side of equation (11) we have constants; consequently, the distribution of τ in our case is uniform. Integrating equation (11) by τ, we obtain theoretical values F_i which are cumulative probability distribution functions of the normal H_0 for values τ_i,

$$F_i = [\tau_i - (-\sqrt{3})] \cdot 0.289 .$$

We calculate the cumulative frequency function, $\tilde{F}_i$, by means of

$$\tilde{F}_i = \frac{i - 0.5}{n} . \quad (12)$$

The values of F_i and $\tilde{F}_i$ and $|F_i - \tilde{F}_i|$ are given in table 7. The probability $P_n(\lambda)$

TABLE 5

AVERAGE VALUES AND STANDARD DEVIATIONS P_2O_5 IN SAMPLES FROM THE GRANITIC ROCKS WITH SiO_2 MORE THAN 60 PER CENT*

No.	Region	$\overline{P_2O_5}$	$S_{P_2O_5}$	n
1	North Finland and Kola peninsula	0.14	0.188	50
2	South and central Finland (on the south from Rovaniemi)	0.17	0.178	141
3	Ukranian Shield	0.14	0.101	119
4	Middle Ural	0.20	0.256	48
5	Northwest Caucasus	0.14	0.099	93
6	Taimyr peninsula and North Land Islands	0.19	0.123	43
7	Central Kazahstan and western Altai	0.15	0.127	173
8	Southwestern Tienshan	0.11	0.086	106
9	Anabara Shield	0.12	0.134	13
10	Watershed region Baikal Lake–Vitim River	0.15	0.142	50
11	Transbaikal	0.14	0.158	107
12	Northeast U.S.S.R. (from the line Vitim–Lena Rivers)	0.08	0.070	51
13	On the Southeast of Amur-Ussuri Rivers	0.15	0.211	33
14	Japan	0.20	0.214	134
15	Eastern China	0.24	0.120	46
16	Indochina	0.12	0.087	42
17	East Indies Islands and Malacca	0.11	0.286	70
18	Australia	0.14	0.142	120
19	British Columbia, Washington, Oregon	0.13	0.068	51
20	Montana, Wyoming, Colorado	0.13	0.123	52
21	California	0.12	0.125	76
22	Canadian Shield	0.19	0.151	56
23	New England and Adirondacks	0.08	0.080	72
24	South America	0.16	0.168	42
25	Greenland	0.32	0.284	25
26	Scotland, North Ireland, and Rockall I.	0.18	0.197	74
27	England and Wales	0.22	0.126	28
28	Portugal	0.40	0.227	26
29	Atlas System	0.25	0.164	49
30	Sahara	0.09	0.081	41
31	Southwestern Africa	0.14	0.079	54
32	Belgian Congo, Angola, Northern Rhodesia	0.15	0.174	52
33	South Africa	0.14	0.132	121
34	Sweden and Denmark	0.18	0.164	83
35	Western Germany	0.28	0.289	64
36	Switzerland	0.20	0.164	222
37	Eastern Germany, Czechoslovakia, Silesia	0.23	0.152	84
38	Madagascar	0.17	0.211	69

* Contents of P_2O_5 are taken from the papers, a list of which will be published later.

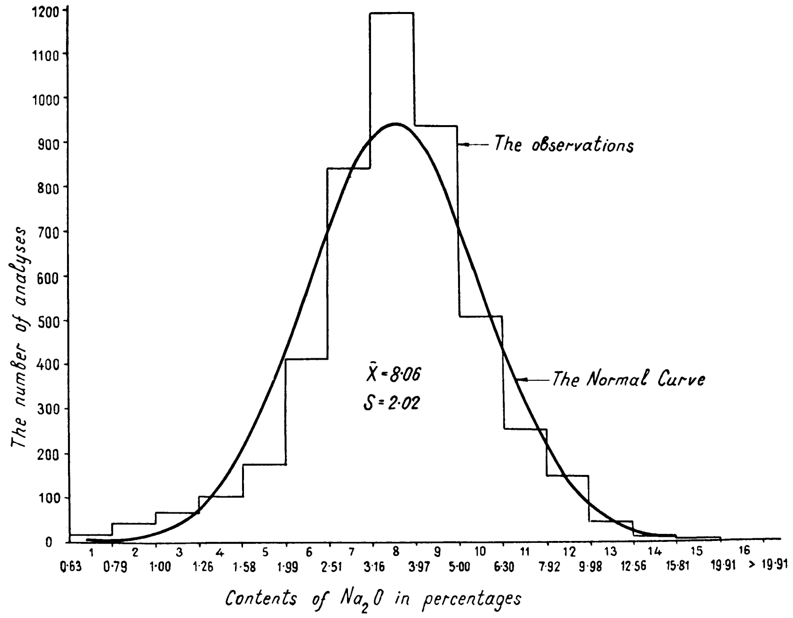

FIG. 3.—Frequency distribution of Na_2O in igneous rocks of the world (logarithmic scale)

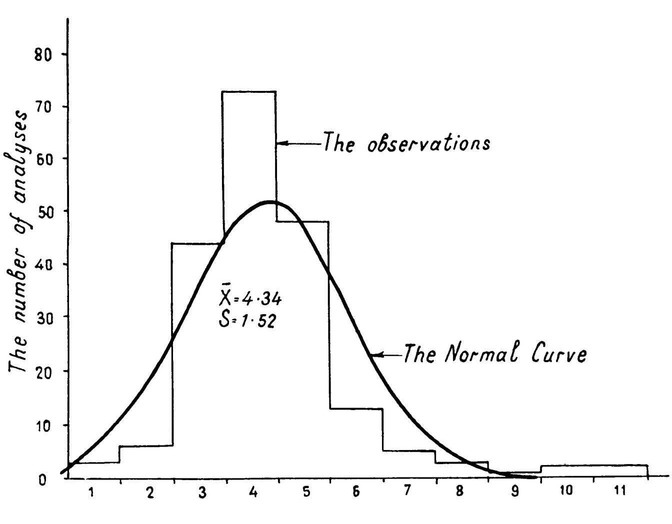

FIG. 4.—Frequency distribution of Na_2O in the basalts of the world (arithmetic scale)

TABLE 6

CONTENTS OF Na_2O IN SAMPLES OF BASALTS

No.	Rocks, Localities, and Authors Cited	Contents of Na_2O in Basalts				$\bar{X}$	$\hat{S}$	$X_i-\bar{X}$	$r_i=(X_i-X)/\hat{S}$
		X_1	X_2	X_3	X_4				
1	2	3	4	5	6	7	8	9	10
1.......	Melilite basalts, S. Africa (Hall, 1938)	2.42	2.03	3.48	3.95	2.970	0.8955	−0.5500	−0.6141
2.......	Basalts, S. Rhodesia (Hall, 1938)	2.38	1.66	1.68	1.94	1.915	0.3352	−0.2550	−0.7607
3.......	Alkali basalts, Kenya (Hall, 1938)	2.63	2.74	2.29	3.05	2.678	0.3078	−0.3880	−1.2605
4.......	Basalts, Madagascar (Hall, 1938)	1.88	2.01	2.19	2.48	2.140	0.2600	+0.3400	+1.3077
5.......	Basalts, Indochina (Lacroix, 1933)	3.15	3.34	3.06	2.65	3.050	0.2912	+0.0100	+0.0343
6.......	Andesite basalts, North China (Lacroix, 1933)	3.31	3.28	3.27	3.28	3.285	0.0173	−0.0050	−0.2890
7.......	Leucitic basalts, Central Java (Van Bemmelen, 1937)	0.73	2.17	4.70	1.85	2.362	1.6773	−1.6320	−0.9730
8.......	Basalts, Aleutian Islands (Coats, 1952)	3.68	3.57	3.65	3.35	3.562	0.1641	+0.0080	+0.0487
9.......	Plagioclase basalts, Sumatra (Westerveld, 1952)	3.43	3.29	3.19	3.47	3.345	0.1291	−0.1550	−1.2006
10.......	Basalts, Krakatau group (Westerveld, 1952)	2.94	2.91	2.46	2.46	2.692	0.2754	−0.2320	−0.8424
11.......	Basalts, Armenia (Struve, 1940)	1.80	3.15	3.55	2.96	2.865	0.7514	+0.6850	+0.9250
12.......	Basalts, Chatanga, Jakutia (Struve, 1940)	2.24	1.91	2.09	1.86	2.025	0.1740	−0.1150	−0.6609
13.......	Basalts, Kamchatka (Struve, 1940)	2.82	2.15	2.33	1.76	2.265	0.4398	+0.5550	+1.2619
14.......	Basalts, Hooker Island, Land of Frantz Josef (Struve, 1940)	2.31	1.80	1.87	0.70	1.670	0.6850	+0.1300	+0.2058
15.......	Alkali basalts, Ussury River, far east of U.S.S.R. (Tatarinov, 1954)	4.03	2.68	2.87	3.16	3.185	0.5969	−0.3150	−0.5277
16.......	Basaltic limburgites, Hilok River, U.S.S.R. (Below, 1956)	3.86	2.67	2.56	3.34	3.108	0.6053	+0.2320	+0.3833
17.......	Basalts, Lower Silesia (Jerzmansky, 1956)	3.27	2.70	2.43	3.64	3.010	0.5469	−0.5800	−1.0605
18.......	Basalts, Australia (Yates, 1954)	2.72	2.98	4.04	3.68	3.355	0.6107	−0.3750	−0.6142
19.......	Basalts, Slovakyan (Šalat, 1955)	2.28	3.23	3.51	2.76	2.945	0.5406	−0.6650	−1.2302
20.......	Basalts, Paricutin, Mexico (Wilcox, 1954)	3.88	3.87	3.84	3.74	3.832	0.0960	+0.0380	+0.3958
21.......	Basalts, Armenia, U.S.S.R. (Zavaritzky, 1953)	2.08	2.16	3.54	2.53	2.578	0.6683	+0.9620	+1.4394
22	Basalts, Avacha, Kamchatka (Gonshakova, 1953)	3.18	2.83	2.45	2.98	2.860	0.3088	+0.1200	+1.3886
23.......	Basalts, Fuji-San, Japan (Tsuya, 1955)	2.67	2.46	2.78	2.65	2.640	0.1331	+0.1400	+1.0518
24.......	Basalts, Hungary (Mauritz and Harwood, 1936)	3.05	3.79	3.28	3.17	3.322	0.3322	+0.4680	+1.4088
25.......	Basalts, Hungary (Jugovics, 1937)	3.02	2.83	5.43	3.48	3.690	1.1917	−0.6700	−0.5623
26.......	Basalts Réunion Island (Lacroix, 1939)	1.41	1.29	0.89	0.80	1.098	0.2955	+0.1920	+0.6498
27.......	Ophitic basalts, California (Powers, 1932)	2.34	2.33	2.24	3.04	2.488	0.3666	−0.2480	−0.6764
28.......	Olivine basalts, Juan Fernández Island (Lacroix, 1928)	2.60	2.94	2.32	3.61	2.868	0.5526	−0.7420	+1.3427
29.......	Basalts, Mangareva Island (Lacroix, 1928)	2.35	2.30	2.31	2.08	2.260	0.1222	+0.0500	+0.4092

TABLE 6—*Continued*

No.	Rocks, Localities, and Authors Cited	Contents of Na_2O in Basalts				$\bar{X}$	$\hat{S}$	$X_i-\bar{X}$	$r_i = (X_i-\bar{X})/\hat{S}$
		X_1	X_2	X_3	X_4				
1	2	3	4	5	6	7	8	9	10
30.......	Taityte, Taity (Lacroix, 1928)	7.83	6.10	8.16	7.16	7.312	0.9145	−1.2120	−1.3253
31.......	Basalts, Mauna Loa, historic age (Macdonald, 1948)	2.43	2.30	2.07	2.29	2.272	0.1591	+0.1580	+0.9931
32.......	Basalts, Kilauea, prehsitoric age (Macdonald, 1948)	2.01	2.42	2.20	2.16	2.198	0.1604	+0.2220	+1.3840
33.......	Basalts, Kilauea, historic age (Macdonald, 1948)	2.64	2.20	2.35	2.45	2.410	0.1848	−0.0600	−0.3247
34.......	Olivine basalts, Hualalai (Macdonald, 1948)	2.30	3.16	3.59	2.89	2.985	0.5401	−0.0950	−0.1759
35.......	Basalts, Mauna Kea (Macdonald, 1948)	2.55	3.08	2.80	2.56	2.748	0.2425	+0.0520	+0.2144
36.......	Basalts, Kohala (Macdonald, 1948)	2.20	3.56	4.45	2.09	3.075	1.1345	+0.4850	+0.4275
37.......	Basalts, Faeroe Islands (Walker and Davidson, 1936)	2.22	2.14	3.89	2.50	2.688	0.8142	−0.4680	−0.5748
38.......	Plagioclase basalts, the Azores (Essenwein, 1929/30)	3.54	2.76	3.24	2.58	3.030	0.4396	−0.2700	−0.6142
39.......	Olivine basalts, the Azores (Essenwein, 1929/30)	2.35	4.17	2.20	2.17	2.722	0.9701	−0.5220	+0.5381
40.......	Basalts, Spitsbergen (Tyrrell and Sanford, 1932/33)	2.49	2.62	2.31	3.18	2.650	0.3756	+0.5300	+1.4110
41.......	Nepheline-melilite basalts, Oahu (Winchell, 1947)	3.93	4.75	4.54	5.12	4.585	0.4981	−0.0450	−0.0903
42.......	Trachybasalts, Jan Mayen (Tyrrell, 1925/26)	3.18	2.37	3.18	3.99	3.180	0.6614	+0.8100	−1.2247
43.......	Basalts, Drakenberg, S. Africa (Walker and Poldervaart, 1949)	1.89	1.74	1.63	1.59	1.712	0.1424	+0.1780	+1.2500
44.......	Doleritic basalts, S. Africa, (Walker and Poldervaart, 1949)	1.92	1.84	1.32	1.89	1.742	0.2878	+0.0980	+0.3405
45.......	Basalts, Mascarene Island (Walker and Nicolaisen, 1954)	2.93	2.48	2.74	3.09	2.810	0.2626	−0.0700	−0.2665
46.......	Olivine basalts (Walker and Nicolaisen, 1954)	3.20	2.25	2.46	2.99	2.725	0.4441	+0.2650	+0.5967
47.......	Doleritic and olivine basalts (Walker and Nicolaisen, 1954)	2.90	3.08	2.77	2.97	2.930	0.1301	−0.1600	−1.2298
48.......	Trachybasalts, Tristan da Cunha and Gough Island (Smith, 1930)	5.84	5.01	7.28	5.25	5.845	1.0182	−0.8350	−0.8201
49.......	Basalts, Japan (Tsuya, 1937)	1.84	2.02	2.47	2.87	2.300	0.4632	−0.4600	−0.9929
50.......	Basalts, West Greenland Noe-Nygaard, 1942)	0.39	1.69	1.01	1.92	1.252	0.6940	+0.4380	+0.6311

TABLE 7

VERIFICATION DATA OF NORMAL H_0 FOR Na_2O

i	τ_i	$\tilde{F}_i$	$\tau_i+\sqrt{3}$	F_i	$\lvert\tilde{F}_i-F_i\rvert$
1	2	3	4	5	6
1........	−1.3253	0.010	+0.4068	+0.117	0.107
2........	−1.2605	0.030	+0.4716	+0.136	0.106
3........	−1.2301	0.050	+0.5020	+0.145	0.095
4........	−1.2298	0.070	+0.5023	+0.145	0.075
5........	−1.2247	0.090	+0.5074	+0.146	0.056
6........	−1.2006	0.110	+0.5315	+0.153	0.043
7........	−1.0605	0.130	+0.6716	+0.194	0.064
8........	−0.9929	0.150	+0.7392	+0.213	0.063
9........	−0.9730	0.170	+0.7591	+0.219	0.049
10........	−0.8424	0.190	+0.8897	+0.257	0.067
11........	−0.8201	0.210	+0.9120	+0.263	0.053
12........	−0.7607	0.230	+0.9714	+0.280	0.050
13........	−0.6764	0.250	+1.0557	+0.305	0.055
14........	−0.0669	0.270	+1.0712	+0.309	0.039
15........	−0.6142	0.290	+1.1179	+0.323	0.033
16........	−0.6142	0.310	+1.1179	+0.323	0.013
17........	−0.6141	0.330	+1.1180	+0.323	0.007
18........	−0.5748	0.350	+1.1573	+0.334	0.016
19........	−0.5623	0.370	+1.1698	+0.338	0.032
20........	−0.5277	0.390	+1.2044	+0.348	0.042
21........	−0.3247	0.410	+1.4074	+0.406	0.004
22........	−0.2890	0.430	+1.4431	+0.417	0.013
23........	−0.2665	0.450	−1.4656	+0.423	0.027
24........	−0.1759	0.470	+1.5562	+0.449	0.021
25........	−0.0903	0.490	+1.6418	+0.474	0.016
26........	+0.0343	0.510	+1.7664	+0.510	0.000
27........	+0.0487	0.530	+1.7808	+0.514	0.016
28........	+0.2058	0.550	+1.9379	+0.559	0.009
29........	+0.2144	0.570	+1.9465	+0.562	0.008
30........	+0.3405	0.590	+2.0726	+0.598	0.008
31........	+0.3833	0.610	+2.1154	+0.611	0.002
32........	+0.3886	0.630	+2.1207	+0.612	0.018
33........	+0.3958	0.650	+2.1279	+0.614	0.036
34........	+0.4092	0.670	+2.1413	+0.618	0.052
35........	+0.4275	0.690	+2.1596	+0.623	0.067
36........	+0.5381	0.710	+2.2702	+0.655	0.055
37........	+0.5967	0.730	+2.3288	+0.672	0.058
38........	+0.6311	0.750	+2.3632	+0.682	0.068
39........	+0.6497	0.770	+2.3818	+0.688	0.082
40........	+0.9250	0.790	+2.6571	+0.767	0.023
41........	+0.9931	0.810	+2.7252	+0.787	0.023
42........	+1.0518	0.830	+2.7839	+0.804	0.026
43........	+1.2500	0.850	+2.9821	+0.861	0.011
44........	+1.2619	0.870	+2.9940	+0.864	0.006
45........	+1.3077	0.890	+3.0398	+0.878	0.012
46........	+1.3427	0.910	+3.0748	+0.888	0.022
47........	+1.3840	0.930	+3.1161	+0.900	0.030
48........	+1.4088	0.950	+3.1409	+0.907	0.043
49........	+1.4110	0.970	+3.1431	+0.907	0.063
50........	+1.4394	0.990	+3.1715	+0.916	0.074

$$\lambda_{50\%} = 0.8280\,; \qquad \frac{\lambda_{50\%}}{\sqrt{50}} \approx 0.117\,; \qquad p\,(\lvert\tilde{F}_j - F_j\rvert > 0.117) > 50\%$$

is approximately equal to $K(\lambda)$ for the inequality

$$\max |\tilde{F}_n - F| < \frac{\lambda}{\sqrt{n}} \qquad (13)$$

and for any λ, $n > 40$,

$$K(\lambda) = 1 - 2\sum_{\nu-1}^{\infty} (-1)^{\nu-1} e^{2\nu^2/2}. \qquad (14)$$

The values of λ_p for some levels of confidence have been tabulated (Dunin-Barkovsky and Smirnov, 1955). We give them in table 8.

The values λ_p which are given in table 8 show that normal H_0 is in agreement with our observation, having a level of confidence of more than 50 per cent, as can be checked by tables 6, 7, and 8 and figure 5.

In this way the question is settled. The frequencies of sodium are distributed normally with a high confidence level for fixed stages of the differentiation of the basalts.

TABLE 8

SHORT TABLE OF PERCENTILE DEVIATIONS FOR FUNCTION $K(\lambda)$

(By Dunin-Barkovsky and Smirnov)

	Per Cent	Per Cent	Per Cent	Per Cent	Per Cent
p......	50	10	5	1	0.1
λ_p.....	0.828	1.224	1.358	1.627	1.950

THE LOCAL DISTRIBUTION

The best material which can be given for the study of the local distribution is a set of the chemical analyses of a small piece of rock. For this reason we took a piece of biotite granodiorite weighing nearly 1 kg., crushed it into 29 small bits of 5 gm. each, and determined with the greatest care the contents of P_2O_5 in them.

The analyzed biotite granodiorite was taken by the author from the outcrop on the right slope of the gorge of the Pjandge River (Pimir) between Pasthoof and Barjumch villages.

The method of determining the P_2O_5 is as follows (Dittler, 1933):

A piece of rock weighing nearly 5 gm. is crushed in an agate mortar and screened through a 100-mesh sieve. The sieved rock is melted with soda in a platinum crucible. The alloy is leached with warm water, filtered off of the insoluble remains, and then these remains are washed by means of a 1 per cent solution of soda. The filtrate is neutralized with nitric acid up to a weak acid reaction and is evapo-

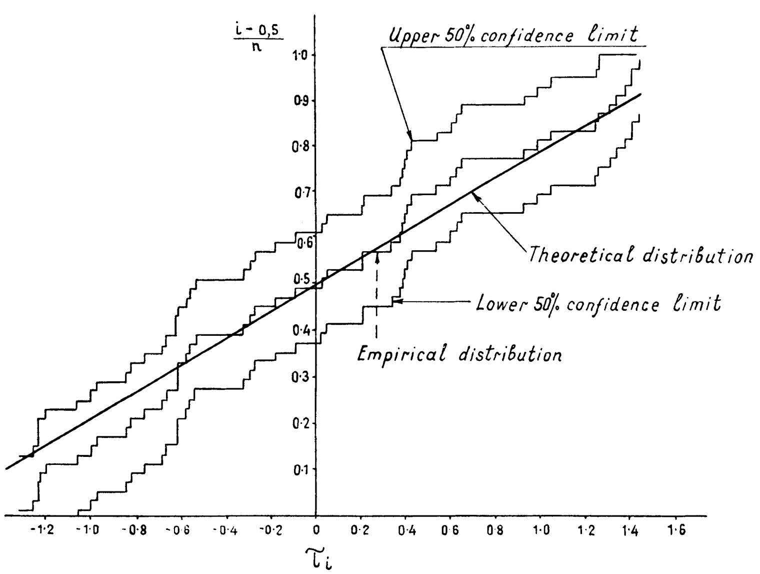

FIG. 5.—Cumulative frequency polygon, its probability distribution function and confidence limits

rated to 50–60 ml. To the solution are added 10 ml. of concentrated nitric acid and 20 ml. of ammonium nitrate (340 gm. NH_4NO_3 in 1l H_2O). The solution is heated up to 50° C., and 25 ml. of $(NH_4)_2M_0O_4$, heated up to 50° C., is added to this solution in the form of a thin squirt, while stirring it with a glass rod (the latter must not touch the walls of the glass). The yellow precipitate of ammonium phosphomolybdate soon separates out.

Identical conditions are strictly kept for precipitations of ammonium phosphomolybdate in all samples (volumes of solutions, their temperatures, the added quantity of reagents, etc.).

When the precipitation is completed, the glass is covered with a watch glass and is left overnight. The next morning the precipitate is filtered through a small dense filter, not shifting the precipitate from the glass to the filter. The precipitate is washed with the help of decantation and then with warm 5 per cent NH_4NO_3 on the filter. The filtrate is mixed and left for the sake of checking the completeness of the precipitation.

Repeated analyses of one and the same piece of rock are given in table 9. From table 9 we see that the errors of determination of P_2O_5 are not greater than 0.01 per cent.

The results of the analysis of the examined 29 pieces of granodiorite are given in table 10.

From figure 6 based on the data of table 10 it is obvious that the local distribution of P_2O_5 in the analyzed biotite granodiorite is normal.

THE FUNDAMENTAL LAW OF THE GEOCHEMICAL PROCESSES

We have stated that the probability distributions of the concentrations of chemical elements in the earth's crust are divided into two groups. The first group of distributions is that of the fixed stage of the geochemical process. These distributions are normal. The second group of distributions is that of mixed products of many stages of the geochemical process. The latter dis-

TABLE 9

DETERMINATIONS OF P_2O_5 REPEATED IN ONE AND THE SAME SAMPLE

(Analyzer, Mrs. N. A. German)

	No. of the Pieces of Granodiorite		
	A	B	C
Determinations of P_2O_5 repeated for one piece..............	0.300; 0.302	0.311; 0.317	0.316; 0.312; 0.314; 0.315

The washed precipitated ammonium phosphomolybdate is dissolved on the filter with warm ammoniac (1:2). The filter is washed several times in warm water with a small amount of ammoniac, and all the water used is collected, together with the solution. The solution and water are collected in an annealed and weighed-up porcelain cup (5.5 cm. in diameter), they are evaporated on a water bath, dried, and carefully annealed with a burner. In the course of annealing, the precipitate changes from white at the beginning to bright yellow and to dark blue at the end of the process ($P_2O_5 \cdot 24MoO_4$). The annealing must be very carefully made up to a temperature not exceeding 400° C. The annealing is ready when all the precipitate becomes dark blue. The cup containing the precipitate is cooled in an exsiccator and then weighed.

TABLE 10

CONTENTS OF P_2O_5 IN SMALL PIECES OF GRANODIORITE FROM PAMIR

(Analyzer, Mrs. N. A. German)

No. of Pieces	Content of P_2O_5 (Per Cent)	No. of Pieces	Content of P_2O_5 (Per Cent)
1......	0.27	16......	0.34
2......	0.29	17......	0.34
3......	0.29	18......	0.34
4......	0.30	19......	0.34
5......	0.30	20......	0.35
6......	0.31	21......	0.35
7......	0.31	22......	0.36
8......	0.31	23......	0.36
9......	0.31	24......	0.36
10......	0.31	25......	0.36
11......	0.32	26......	0.37
12......	0.32	27......	0.38
13......	0.32	28......	0.38
14......	0.33	29......	0.39
15......	0.33	....	....

tributions are skew, with a large positive skewness. Our analysis of geochemical peculiarities, by means of which we obtain the local and joint distributions, gives us the possibility of finding the most general features of the geochemical processes. We have named these features the "fundamental law of the geochemical processes,"[6] the fundamental law of the geochemical processes being as follows: The joint probability distribution function of the concentration of the minor chemical element deposited by natural chemical reactions has a large positive skewness. This skewness indicates that the deposition of small concentrations of the minor element by geochemical processes is, as a rule, more stable than the deposition of large concentrations of this element by the same geochemical process—the quantity of the deposited element in the form of small concentrations being not less than that in the form of large concentrations. It seems to us that prospecting and mining confirm our statements.

We assume that the fundamental law of the geochemical processes has independent importance and has no relation to Goldschmidt's point of view. The fact is that when Goldschmidt (1954) wrote: "Modern geochemistry studies the distribution and amount of the chemical elements in minerals, ores, rocks, soils, waters, and the atmosphere, and the circulation of the elements in nature, on the basis of the properties of their atoms and ions" he, as well as all other classicists of geochemistry such as V. I. Vernadsky, F. E. Clarke, A. E. Fersman, meant a deterministic approach to nature. In the cited papers published by Ahrens as well as the papers of the author, including the present one, we have the probabilistic approach to nature. Goldschmidt and Vernadsky spoke about concrete concentrations of the chemical elements, whereas we speak about probability distributions of the concentrations. These are quite different things. For instance, let us take two flakes of mica. In the first flake we have 0.1 per cent Rb_2O, in the second one we have 0.2 per cent Rb_2O, with normal probability distributions in both flakes. From Goldschmidt's point of view, the distributions of Rb_2O_0 in the flakes are different because in one of them the concentration of Rb_2O is

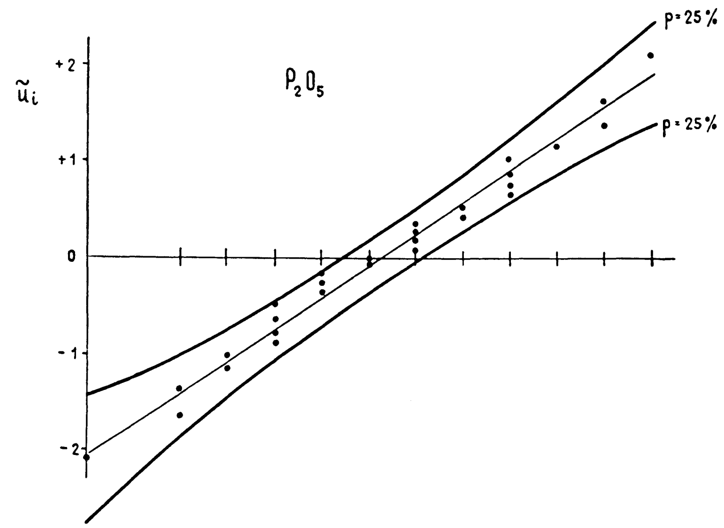

FIG. 6.—The straight-line diagram and confidence limits for the distribution of P_2O_5 in the piece of granodiorite from Pamir.

[6] We did not include major elements or mechanical deposits.

twice as great as that in the second. From our point of view, the analyzed flakes have similar normal probability distributions of the concentrations of Rb_2O. Further, let us take two pieces of feldspar, one of which has a normal and the second a lognormal probability distribution of the concentration of Cs_2O, with average concentrations of Cs_2O in every piece equaling 0.01 per cent; From Goldschmidt's point of view the pieces have equal distributions of Cs_2O, because in every piece it equals 0.01 per cent; from our point of view the probability distributions in the analyzed pieces are different because one is normal and the other is lognormal; by means of the analysis of the stochastic sense of the probability distributions of the concentrations of Cs_2O in pieces, we can understand the difference between the processes of the concentration of Cs_2O in the minerals of the pieces analyzed.

In modern science we deal with the increasing role of the probabilistic approach to nature. The formulation of the fundamental law of geochemical processes is the reflection of modern tendencies in science.

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