



Modeling Particle Size Distribution in Lunar Regolith via a Central Limit Theorem for Random Sums

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Abstract: A version of the central limit theorem is proved for sums with a random number of independent and not necessarily identically distributed random variables in the double array limit scheme. It is demonstrated that arbitrary normal mixtures appear as the limit distribution. This result is used to substantiate the log-normal finite mixture approximations for the particle size distributions of the lunar regolith. This model is used as the theoretical background of the two different statistical procedures for processing real data based on bootstrap and minimum χ^2 estimates. It is shown that the cluster analysis of the parameters of the proposed models can be a promising tool for revealing the structure of such real data, taking into account the physico-chemical interpretation of the results. Similar methods can be successfully used for solving problems from other subject fields with grouped observations, and only some characteristic points of the empirical distribution function are given.

Keywords: central limit theorem; random sequence; random index; transfer theorem; random sum; random Lindeberg condition; log-normal mixtures; grouped data; bootstrap; minimizing χ^2 estimate; EM algorithm; statistical estimations

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1. Introduction

The study of regularities in the sizes of particles of lunar and other planetary regoliths is relevant for planning and design of manned and unmanned space missions for exploration of space bodies such as the Moon, asteroids, planets, and their satellites. In the conditions of low gravity and the absence of dense atmosphere, dust structures acquire properties that are not typical for Earth conditions: they form clouds of charged particles that have very high abrasive properties. They deposit on structural elements of stations and spaceships, e.g., on solar batteries, and space suites causing their malfunction and failures. Thus, the solution of the problem under consideration may turn very useful and prospective in the preparation of new space missions to increase their safety and general success.

In the present paper, we consider the problem of modeling the distribution of the sizes of dust particles in the lunar regolith formed as a result of multiple impacts, e.g., of the bombardment of the lunar surface by meteorites. These impacts are accompanied by both processes of explosion dispersion of particles with their fragmentation and processes of the agglomeration of particles in exothermic plasma-chemical synthesis reactions [1]. As it was noted in the book [2], the observed statistical distributions of the particle size in lunar regolith, as a rule, have heavy power-type tails.

In this paper, we combine and generalize the results of the papers [3,4] in order to obtain a general model of the particle size distribution that can be applied to describe statistical regularities in the lunar regolith probes brought by the Apollo missions. The proposed models have the form of asymptotic approximations obtained by a central limit theorem for sums of a random number of independent not necessarily identically distributed random variables in the special double array limit scheme.

Some words about the history of the problem. Initially, the studies of these models concerned pure fragmentation of particles related to mining processes. In the paper [5] by N. K. Razumovsky published in 1940, many cases were described where the logarithms of the particle sizes (e.g., of gold grains or ore particles under crushing) had approximately normal (Gaussian) distribution. This paper attracted the attention of A. N. Kolmogorov who suggested a mathematical model that analytically explained the appearance of the log-normal distribution of the particle size under crushing, as well as minerals in geological samples [6]. The mathematical model suggested by Kolmogorov was based on the study of the change of the number of particles, whose size does not exceed a specified threshold, in time. Kolmogorov's result is valid under rather strong assumptions. In particular, within the framework of Kolmogorov's model, for the distribution of particle size under crushing to be log-normal, it is necessary that the crushing rate is constant, that is, does not depend on the current sizes of the crushed particles. At the same time, it is practically obvious that, as the particle (grain) becomes smaller, the intensity of its collisions with other particles (grais) or elements of the crusher may change, e.g., decrease by virtue of the fact that the probability of a collision of the particle with other particles is in some sense proportional to the particle size so that, the smaller the particle, the less is this probability. This circumstance was noted by Kolmogorov himself, who wrote in the final part of his paper [6]: "It would be interesting to study mathematical schemes in which the crushing rate decreases (or increases) with the decrease of their sizes. As this is so, first of all, it is natural to consider the cases where the crushing rate is proportional to some power of the particle size. If this power differs from zero, then, apparently, the log-normal law will not be applicable". In the paper [7], the results of statistical analysis of real data were presented that confirmed the log-normal model for the distribution of the sizes of grains in rock formations.

However, some works contain empirical evidence that sometimes the log-normal model for the distribution of particle size under crushing is not adequate. As far back ago as in 1954, R. A. Bagnold in his book [8] noticed that the logarithm of the probability density function of the logarithm of the particle size in natural sand deposits resembles a hyperbola rather than a parabola. This means that, in accordance with Bagnold's observations, the particle size distribution possesses exponentially decreasing tails rather than log-normal.

This circumstance induced some specialists to pay attention to such models as log-asymmetric Laplace distribution, the so-called two-sided Pareto-log-normal distribution or log-normal//inverse Gaussian (NIG) distribution. In particular, in [9,10], the models were considered for the process of forming of the particle size distribution under crushing based on the assumption that, while moving from one place to another, a particle can be split into some smaller particles as a result of collisions or other impacts implying the randomness of the particle mass after splitting. The particles do not reach the terminal point simultaneously. Some of them can change the direction of their motion or stick for a long time while others can pass their way much sooner without collisions. The state of a particle obviously depends on its distance from the terminal point. The Reed–Jorgensen and Sørensen models take this realities into account by assuming that the time the particle is observed at is random. Actually, Reed, Jorgensen and Sørensen introduced randomization into the model suggested by Kolmogorov in 1941, by assuming that it takes different times for different particles to move from one source to another.

In the Reed–Jorgensen model, the time from the start of the particle's motion to its "termination" is assumed to be an exponentially distributed random variable, so that the final particle size distribution turns out to be two-sided Pareto-log-normal.

Probability distributions for which the logarithm of the density function is a hyperbola (hyperbolic distributions) were introduced by O.-E. Barndorff-Nielsen in 1977 [11]. In several papers, it was noted that hyperbolic distributions demonstrated high concordance with empirical distributions of real geological data; see, e.g., [12–14]. Actually, in Ref. [11], a more general class of probability distributions was introduced, the class of so-called generalized hyperbolic distributions. The NIG distribution is a representative of this class. By definition, this distribution is a special (variance-mean) mixture of normal distribution, the mixing distributions of the logarithm of particle size than the hyperbolic distribution. This is due to that the tails of NIG distributions are heavier than those of the "ordinary" hyperbolic distribution, as is typical, say, for the natural sand deposits. Another advantage of the NIG distributions is that they possess many favorable mathematical properties. For example, their moments can be calculated explicitly.

In the Sørensen model, the motion of the particle is assumed Brownian. Therefore, the time from the start of the motion to its termination is random with the inverse Gaussian distribution. This results in the logarithm of the particle size having the NIG distribution.

The Reed–Jorgensen and Sørensen models demonstrate very good fit to the empirical data of particle (grain) sizes in natural deposits. However, these models do not take into account Kolmogorov's remark: in these models, the crushing intensity remains constant.

In Ref. [3], a more general model was considered within which the crushing intensity could be variable and even random. In [3], the process of relative changes of the particle size was assumed to be compound doubly stochastic Poisson process, and it was demonstrated that the randomness of the intensity of the crushing process might lead to the particle size distribution having the form of a mixture of log-normal distributions. In accordance with such a model, the distribution of the number of size changes prior to any observation time is mixed Poisson. As it was noted in [15], these processes are natural models of non-stationary chaotic stochastic processes possessing some extremal entropy properties. It should be mentioned that in the Reed–Jorgensen and Sørensen models, a homogeneous Poisson point process was considered as a model of the flow of events implying the changes of the particle size.

As it has been already noted, in all the works mentioned above, the process of pure fragmentation of particles was considered so that every impact of particles could result in a decrease of their sizes only, whereas, in plasma-chemical reactions in the regoliths on the surface of space bodies devoid of dense atmospheres, every impact can result in both decrease and increase of the particle size, say, due to sintering. In the present paper, we consider models for the particle size distribution that take this circumstance into account. Unlike Kolmogorov's approach where the main object of study was the number of particles with a certain size, here we consider the evolution of the size of a separate particle.

One more characteristic feature of the models considered in the present paper is their generality. All the models mentioned above have the form of special mixtures of log-normal distributions: the Reed–Jorgensen, Sørensen, and Barndorff–Nielsen models have the form of normal variance–mean mixtures. In these mixtures, the scale and location parameters of the mixed distributions are rigidly bound so that actually mixing is performed with respect to one parameter. This construction noticeably complicates the physical interpretation of the obtained models. In the present paper, we propose a rather realistic scheme of forming of models that are general mixtures of log-normal models in which the mixing distribution is, in general, two-variate. Although these models seem to be too general so that the statistical estimation of the distributions can be very complicated, they allow natural discrete approximations for the mixing distributions. In this case, the approximate model for the particle size distribution is a finite mixture of log-normal distributions, whose parameters can be estimated by standard statistical procedures of the EM-algorithm type.

The paper is organized in the following way. Section 2 contains the description of the mathematical foundation of general normal mixture models for the particle size distribution. We formulate a new general version of the central limit theorem for random sums in a special double array limit scheme

stating that, under rather general and reasonable random Lindeberg condition, the distribution of the sum of a random number of independent random variables with finite second moments approaches the set of normal location-scale mixtures, so that, within this set, a mixture can be chosen that can be used as an asymptotic approximation to the distribution of the random sum. This result is used in Section 3 as the base for the main model of the particle size distribution. In Section 4, we discuss the approximation of the sample distribution function constructed from data given in the NASA catalog [16] by the method of statistical simulation of samples by the bootstrap technique. In Section 5, we discuss the approximation of the sample distribution function by the minimization of the χ^2 statistic. In Section 6, we use the cluster analysis to separate some typical regularities in the parameters of distributions fitted to all 317 samples of lunar regolith presented in the NASA catalog [16]. The obtained results demonstrate that the finite normal mixture model fits the real data very well measured in both the classical metric scale (micrometers, μ m) and ϕ scale conventional in geology and selenology [17]. Appendix A contains the description of the algorithms used for the approximation of the distribution of

2. Central Limit Theorem for Sums of a Random Number of Independent Not Identically Distributed Random Variables in the Special Double Array Limit Scheme

application of proposed algorithms for lunar regolith samples.

Assume that all the random variables considered in this paper are defined on one and the same probability space $(\Omega, \mathfrak{F}, \mathbb{P})$. The symbols $\stackrel{d}{=}$ and \implies will denote coincidence of distributions and weak convergence (convergence in distribution).

particle size in lunar regolith by finite normal mixtures. Appendix B presents figures that demonstrate

Let $n, k \in \mathbb{N}$ and $\{S_{n,k}\}$ be a double array of random variables, $a_{n,k}$ and $b_{n,k} > 0$ are real numbers, $n, k \in \mathbb{N}$. The row-wise independence of $\{S_{n,k}\}_{k \ge 1}$ is not assumed. Consider a family of the random variables

$$\left\{Y_{n,k} \equiv \frac{S_{n,k} - a_{n,k}}{b_{n,k}}\right\}_{n,k \in \mathbb{N}} \tag{1}$$

with characteristic functions $f_{Y_{n,k}}(t)$, $t \in \mathbb{R}$. Consider a family $\{N_n\}_{n \in \mathbb{N}}$ of nonnegative integer-valued random variables such that, for each $n, k \in \mathbb{N}$, the random variables N_n and $S_{n,k}$ are independent. Let Y be a random variable with distribution and characteristic function $F_Y(x)$ and $f_Y(t), x, t \in \mathbb{R}$, respectively.

Definition 1 ([4]). The random variables $Y_{n,k}$ and Y are said to satisfy the coherency condition, if, for any $T \in (0, \infty)$,

$$\lim_{n \to \infty} \mathbb{E} \sup_{|t| \leq T} \left| f_{Y_{n,N_n}}(t) - f_Y(t) \right| = 0.$$
⁽²⁾

It is worth noting that, if row-wise convergence $Y_{n,k}$ to Y takes place, the relation

$$\lim_{k \to \infty} \sup_{|t| \le T} |f_{Y_{n,k}}(t) - f_Y(t)| = 0.$$
(3)

holds for any $n \in \mathbb{N}$ and $T \ge 0$. Thus, we can say that coherency condition (2) means that "pure" row-wise convergence (3) takes place "on the average" so that the "row-wise" convergence as $k \to \infty$ is somehow coherent with the "principal" convergence as $n \to \infty$. Following [4], below we will deal with the case where the random variables $S_{n,k}$ have the additive structure and are formed as sums of independent random variables with the finite second moment. It turns out that, under these additional "structural" assumptions, the role of coherency condition is played by the "random Lindeberg condition". Furthermore, if we deal not with a double array of random variables but simply with a sequence of random variables, then, as was shown in [18,19], the coherency condition turns out to be equivalent to the following set of conditions: the indices N_n infinitely increase in probability that is $\mathbb{P}(N_n \leq m) \to 0$ as $n \to \infty$ for any $m \ge 0$, and either $b_n \to 0$ or $b_n \to \infty$ as $n \to \infty$.

Let c_n and $d_n > 0$ be real numbers, $n \in \mathbb{N}$. Denote

$$U_n = \frac{b_{n,N_n}}{d_n}, \quad V_n = \frac{a_{n,N_n} - c_n}{d_n}, \quad Z_n = \frac{S_{n,N_n} - c_n}{d_n}.$$

Let $\{X_{n,j}\}_{j \ge 1}$, $n \in \mathbb{N}$, be a double array of row-wise independent not necessarily identically distributed random variables with distribution functions $F_{n,j}(x)$. Denote

$$S_{n,k} = X_{n,1} + \dots + X_{n,k}, \quad n,k \in \mathbb{N},$$
 (4)

The following statement is actually a transfer theorem for the double array limit scheme under consideration.

Lemma 1 ([4]). Assume that coherency condition (2) holds. If there exist random variables U and V such that the joint distributions of the couples (U_n, V_n) converge to that of the couple $(U, V) : (U_n, V_n) \Longrightarrow (U, V)$ as $n \to \infty$, then

$$Z_n \Longrightarrow Z \stackrel{d}{=} Y \cdot U + V \tag{5}$$

as $n \to \infty$, where the random variable Y is independent of the couple (U, V).

It follows from Lemma 1 that the distribution and characteristic functions of random variables Z and Y are related as

$$F_Z(x) = \mathbb{E}F_Y\left(\frac{x-V}{U}\right), \quad f_Z(t) = \mathbb{E}f_Y(tU)e^{itV}, \quad x \in \mathbb{R}, \ t \in \mathbb{R}.$$
(6)

that is, the limit law for normalized randomly indexed random variables Z_n is a scale-location mixture of the distributions, which are limiting for normalized non-randomly indexed random variables $Y_{n,k}$.

For fixed random variables *Z* and *Y*, introduce the set W(Z|Y) containing all couples of random variables (U, V) such that the characteristic function $f_Z(t)$ can be represented in the form (6) and $\mathbb{P}(U \ge 0) = 1$. Whatever random variables *Z* and *Y* are, the set W(Z|Y) is always nonempty since it trivially contains the couple (0, Z). The set W(Z|Y) may contain more than one element. For example, if *Y* is the standard normal random variable and $Z \stackrel{d}{=} W_1 - W_2$ where W_1 and W_2 are independent random variables with the same standard exponential distribution, then, along with the couple $(0, W_1 - W_2)$, the set W(Z|Y) contains the couple $(\sqrt{W_1}, 0)$. In this case, $F_Z(x)$ is the symmetric Laplace distribution.

Definition 2 ([20]). A family of random variables $\{X_j\}_{j \in \mathbb{N}}$ is said to be weakly relatively compact, if each sequence of its elements contains a weakly convergent subsequence. In the finite-dimensional case, the weak relative compactness of a family $\{X_j\}_{j \in \mathbb{N}}$ is equivalent to its tightness:

$$\lim_{R\to\infty}\sup_{n\in\mathbb{N}}\mathbb{P}(|X_n|>R)=0$$

Let $L_2((X_1, X_2), (Y_1, Y_2))$ be any probability metric which metrizes weak convergence in the space of two-dimensional random vectors, for example, the Lévy–Prokhorov metric [21].

Lemma 2 ([4]). Assume that the random variables $S_{n,k}$ have the form (4). Let the family of random variables $\{Y_{n,k}\}_{n,k\in\mathbb{N}}$ be weakly relatively compact. Assume that coherency condition (2) holds. Then, convergence (5) of normalized random sums Z_n to some random variable Z takes place with some $c_n \in \mathbb{R}$ if and only if there exists a weakly relatively compact sequence of couples $(U'_n, V'_n) \in W(Z|Y)$, $n \in \mathbb{N}$, such that condition

$$\lim_{n \to \infty} L_2\left((U_n, V_n), (U'_n, V'_n) \right) = 0$$
(7)

holds.

Denote $\mu_{n,j} = \mathbb{E}X_{n,j}$, $\sigma_{n,j}^2 = \mathbb{D}X_{n,j}$ and assume that $0 < \sigma_{n,j}^2 < \infty$, $n, j \in \mathbb{N}$. Denote

$$A_{n,k} = \mu_{n,1} + \dots + \mu_{n,k} \ (= \mathbb{E}S_{n,k}), \quad B_{n,k}^2 = \sigma_{n,1}^2 + \dots + \sigma_{n,k}^2 \ (= \mathbb{D}S_{n,k})$$

It is easy to make sure that $\mathbb{E}S_{n,N_n} = \mathbb{E}A_{n,N_n}$, $\mathbb{D}S_{n,N_n} = \mathbb{E}B_{n,N_n}^2 + \mathbb{D}A_{n,N_n}$, $n \in \mathbb{N}$. In order to formulate a version of the central limit theorem for random sums with the limiting distribution being an arbitrary normal mixture, assume that non-random sums, as usual, are centered by their expectations and normalized by by their mean square deviations and put $a_{n,k} = A_{n,k}$, $b_{n,k}^2 = B_{n,k}^2$, $n, k \in \mathbb{N}$. As this is so, as c_n and d_n^2 we can (but are not obliged to) take $\mathbb{E}A_{n,N_n}$ and $\mathbb{E}B_{n,N_n}^2 + \mathbb{D}A_{n,N_n}$, respectively.

Now let Y be a random variable with the standard normal distribution:

$$\mathbb{P}(Y < x) = \Phi(x) \equiv \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{x} e^{-z^2/2} dz, \quad x \in \mathbb{R}.$$

Theorem 1. Assume that the random Lindeberg condition holds: for any $\varepsilon > 0$,

$$\lim_{n \to \infty} \mathbb{E} \frac{1}{B_{n,N_n}^2} \sum_{j=1}^{N_n} \int_{|x-\mu_{n,j}| > \varepsilon B_{n,N_n}} (x-\mu_{n,j})^2 dF_{n,j}(x) = 0.$$
(8)

Then, the convergence (5) of random variables Z_n to Z as $n \to \infty$ takes place with some $c_n \in \mathbb{R}$ and $d_n > 0$ if and only if there exists a weakly relatively compact sequence of couples $(U'_n, V'_n) \in W(Z|Y)$, $n \in \mathbb{N}$, such that, for any $n \in \mathbb{N}$ relation, (7) holds and

$$\mathbb{P}(Z < x) = \mathbb{E}\Phi\left(\frac{x - V'_n}{U'_n}\right), \quad x \in \mathbb{R},$$
(9)

Proof. We will deduce Theorem 1 from Lemma 2. First, check that, in the case under consideration, the family of random variables $\{Y_{n,k}\}_{n,k\in\mathbb{N}}$ is weakly relatively compact. For R > 0, by the Chebyshev inequality, we have

$$\mathbb{P}\left(|Y_{n,k}| > R\right) = \mathbb{P}\left(\left|\frac{S_{n,k} - A_{n,k}}{B_{n,k}}\right| > R\right) \leqslant \frac{1}{R^2} \longrightarrow 0$$

as $R \to \infty$, that is, the family $\{Y_{n,k}\}_{n,k \in \mathbb{N}}$ is tight and hence weakly relatively compact.

Second, exactly in the same way as it was done in the proof of Theorem 4 in [4], it can be proved that the random Lindeberg condition (8) implies the coherency condition (2) with $f_Y(t)$ being the characteristic function of the standard normal distribution, $f_Y(t) = e^{-t^2/2}$, $t \in \mathbb{R}$.

Thus, Theorem 1 becomes a simple consequence of Lemma 2. \Box

Actually, Theorem 1 means that, under a rather general and reasonable random Lindeberg condition, the distribution of the sum of a random number of independent random variables with finite second moments approaches the set of normal location-scale mixtures, so that, with finite n, within this set, a mixture can be chosen that can be used as an asymptotic approximation to the distribution of the random sum.

Theorem 1 is a substantial generalization and strengthening of a result of [22] where a special version of the random Lindeberg condition was used with geometrically distributed random variables N_n to establish sufficient conditions of convergence of the distributions of random sums to the Laplace law. In turn, Ref. [22] was not the first work where the random Lindeberg condition was used. Apparently, it was introduced in [23], where several equivalent formulations of this condition were presented. In [24], it was proved that, under some additional assumptions, the random Lindeberg condition is necessary and sufficient for the convergence of the distributions of randomly centered

random sums to the normal distribution. Here, we consider a case significantly more general than those considered in [24] or [22].

Since, unlike one-parameter normal mixtures (pure scale mixtures, pure location mixtures, variance–mean mixtures), two-parameter normal scale-location mixtures are not identifiable, in Theorem 1 condition (7) describing the rapprochement of the sequence of couples of the random variables $\{(U_n, V_n)\}_{n \ge 1}$ with the special set of couples in Theorem 1 cannot be replaced by a condition of convergence of this sequence to an element of this set so that the 'if and only if' character of Theorem 1 was preserved. However, the latter condition (which is obviously stronger than (8)) together with the random Lindeberg condition are sufficient for the convergence (5). Namely, the following transfer theorem holds concretizing Lemma 1.

Corollary 1. Assume that the random Lindeberg condition (8) holds and there exist $c_n \in \mathbb{R}$, $d_n > 0$ and a couple of random variables (U, V) such that, as $n \to \infty$

$$\left(\frac{B_{n,N_n}}{d_n},\frac{A_{n,N_n}-c_n}{d_n}\right) \Longrightarrow (U,V)$$

Then, as $n \to \infty$ *,*

$$\mathbb{P}\left(\frac{S_{n,N_n}-c_n}{d_n}< x\right) \Longrightarrow \mathbb{E}\Phi\left(\frac{x-V}{U}\right).$$

For $n, j, k \in \mathbb{N}$, denote $v_{n,j}^3 = \mathbb{E}|X_{n,j} - \mu_{n,j}|^3$, $M_{n,k}^3 = v_{n,1}^3 + \ldots + v_{n,k}^3$, $L_{n,k}^3 = M_{n,k}^3 B_{n,k}^{-3}$. In [4], it was demonstrated that, if the third absolute moments of the summands exist, then the random Lindeberg condition (8) follows from the *random Lyapunov condition*

$$\lim_{n\to\infty}\mathbb{E}L^3_{n,N_n}=0$$

which seems to be more easily verifiable than the random Lindeberg condition.

3. Mixed Log-Normal Models for the Distribution of the Particle Size

Although in this paper we will follow the general reasoning of [3], here we will use a scheme that is more general than that considered in [3]. The initial size of a particle (say, at some time instant t_0 to which, for convenience, we will assign that the value $t_0 = 0$) will be denoted s_0 . As time goes by, the particle undergoes collisions with other particles or other transformations due to heating, impacts with meteoroids, etc. We assume that the *i*th collision (transformation) results in the fact that, after it, the particle size becomes $D_i > 0$ times that of its size before the *i*th transformation. Let N(t) be the number of transformations of the particle by time *t*. Then, the particle size at time *t* has the form $Z(t) = s_0 \prod_{i=1}^{N(t)} D_i$. Hence,

$$S(t) \equiv \log Z(t) = \mu + \sum_{i=1}^{N(t)} X_i,$$
(10)

where $\mu = \ln(s_0)$ and $X_i = \ln D_i$.

In [3], it was assumed that $X_1, X_2, ...$ are independent and identically distributed with $\sigma^2 \equiv \mathbb{D}X_i < \infty$. The assumptions of the coincidence of the distributions of $X_1, X_2, ...$ are too restrictive since different collisions or transformations may be caused by processes of different types, say, fragmentation or agglomeration. Instead, here we will not impose such a condition.

As a theoretical foundation of the models proposed here, we will use the limit scheme that was considered in the preceding section and for the sake of generality introduce an auxiliary formal parameter $n \in \mathbb{N}$ and assume that, instead of one sequence of independent random variables X_1, X_2, \ldots , we will consider a double array $\{X_{n,j}\}_{j\geq 1}, n \in \mathbb{N}$, of row-wise independent not necessarily identically distributed random variables. As above, we use the notation $S_{n,k}$ for the sum $S_{n,k} = X_{n,1} + \ldots + X_{n,k}$, $n, k \in \mathbb{N}$.

Instead of a single counting process N(t), now we will have a sequence of counting processes $N_n(t)$, so, instead of basic structural model (9), we will deal with the more general model $S_n(t) = \mu_n + S_{n,N_n(t)}$. Fix the 'terminal' time t = T > 0 and assume that $N_n = N_n(T)$ for any $n \ge 1$ so that, by definition, $S_n(T) = S_n$ and

$$S_n = \mu_n + S_{n,N_n}.\tag{11}$$

Model (11) can be interpreted in the following way. For a fixed n, the sequence $X_{n,1}, \ldots, X_{n,N_n}$ describes a possible trajectory of the process of changes of the particle size. Actually, we are interested in the asymptotic approximation for the distribution of the 'terminal' size S_{n,N_n} (4) with large N_n (with large T or with a large intensity of transformations). Having introduced the auxiliary (infinitely large) parameter n, we obtained the possibility to consider the scheme within which it is admitted that, as n changes, the distributions of $X_{n,j}$ can also change as if, instead of a single particle, we deal with a set of particles with 'long and possibly different histories'. As this is so, each trajectory starts with its own initial size $s_{n,0}$ so that $\mu_n = \ln s_{n,0}$

As in Section 2, denote $\mu_{n,j} = \mathbb{E}X_{n,j}$, $\sigma_{n,j}^2 = \mathbb{D}X_{n,j}$ and assume that $0 < \sigma_{n,j}^2 < \infty$, $n, j \in \mathbb{N}$. In all the preceding papers, pure fragmentation (crushing) was considered, and it was assumed that $0 \leq D_j \leq 1$ resulting in $\mu = \mathbb{E}X_1 = \mathbb{E}\log D_i \leq 0$. The equality of μ to zero corresponded to the case of 'degenerate' fragmentation leaving each particle unchanged. In the present paper, the parameters $\mu_{n,j}$ can be both positive and negative. The case $\mu_{n,j} < 0$ corresponds to fragmentation, whereas the case $\mu_{n,j} > 0$ corresponds to agglomeration.

The assumption $0 < \sigma_{n,j}^2 < \infty$ is not superfluously restrictive. From the practical point of view, this assumption actually withdraws beyond the range of considered cases only those situations that admit either instantaneous zeroing of the particle size or, vice versa, instantaneous increase of the size of a small particle to very large (infinitely large) values. As one more argument in favor of the assumption of finiteness of the second moment of the random variables $X_{n,j}$, note that stable laws are well-known examples of distributions with very heavy tails decreasing as power functions with a small exponent. However, even their logarithmic moments are finite.

Based on Theorem 1, we can conclude that, under very general conditions, the approximation to the distribution of the logarithm of the particle size should be sought within the family of normal scale-location mixtures. In other words, the approximation to the distributions of the particle size itself should be sought within log-[normal scale-location mixtures,] that is, if *Z* is the particle size, then

$$\mathbb{P}(Z < x) = \mathbb{P}(\log Z < \log x) \approx \mathbb{E}\Phi\left(\frac{\log x - V}{U}\right)$$
(12)

for some couple of random variables $(U, V) \in W(Z|X)$. It can be easily seen that the log-[normal scale-location mixture] on the right-hand side of (12) is actually the scale-location mixture of log-normal distributions.

Furthermore, from the definition of the Lebesgue integral, it follows that any normal scale-location mixture can be approximated by a finite normal scale-location mixture, that is, in practice, the approximations to the distribution of the particle size can be sought in the form

$$\mathbb{P}(Z < x) = \sum_{i=1}^{k} p_i \Phi\left(\frac{\log x - a_i}{\sigma_i}\right)$$
(13)

with some $K \in \mathbb{N}$, $p_k \ge 0$, $v_k \in \mathbb{R}$, $u_k \ge 0$, k = 1, ..., K, $p_1 + ... + p_K = 1$. Finite normal mixture model (13) is more convenient and in some sense more interpretable since it allows for naturally separating some typical regularities in the data by well-known methods of statistical analysis, say, by the EM algorithm. It should be noted that the approximation of the general normal mixture by a finite mixture (13) is a kind of regularization of the originally ill-posed problem of estimation of the parameters of an unidentifiable general normal mixture, whereas finite normal mixtures are identifiable, that is, the problem of estimation of their parameters has a unique solution [25].

4. Approximation of a Sample Distribution Function Based on Sample Simulation

In the NASA catalog [16], the data are presented in the tabular form as the pairs "particle size–percentage of particles of this size in the sieved samples". Information is available only on a few (as a rule, no more than ten) points of growth (selected, generally speaking, haphazardly) of the empirical distribution function, but there are no data between these points. In the original paper [16], Steineman's interpolation [26] was used to approximate the distributions, and then the corresponding histograms were built using the macros of the Excel.

In this paper, piecewise cubic Hermite interpolating polynomials (see, for example, [27]) are used for similar purposes. They allowed us to obtain the curves that are closest to those presented in the NASA catalog.

The proposed approximation by Hermite polynomials made it possible to obtain a continuous empirical distribution function (ECDF) that allows for using the method of inverse transform sampling. The sample size for the estimation of the parameters was 10,000 observations. In addition, to test the homogeneity hypotheses concerning the coincidence of the approximating mixture and the original empirical distribution function by the one-sample Kolmogorov–Smirnov goodness-of-fit test, one more independent sample of 2500 observations was generated. In general, this procedure is methodically close to bootstrap. Its description is given in Algorithm A1.

The generated samples are used to obtain maximum likelihood estimates for the parameters of the approximating distribution (13) by the EM-algorithm for finite normal moxtures [28]. Some examples of applying this bootstrap procedure to real samples of the lunar regolith are shown on Figures A1–A9.

The graphs on the left show the initial data obtained in the sifting process from the tables of the NASA catalog (squares), their interpolation using Hermite polynomials (blue solid line), and the approximating mixture (magenta line). It is seen that both curves coincide almost everywhere. This situation is typical for the vast majority of analyzed samples. It is worth noting that several values were checked for the number *k* of components in (13). It was empirically found that the necessary balance between the quality of the approximation and the computational complexity is achieved at the value k = 4, which was further used to process all 317 samples.

The graphs on the right on Figures A1–A9 demonstrate the histograms for the simulated samples and sifted data constructed from ECDF for a particular sample, as well as their approximation by the density of the mixture distribution. It should be noted that the accuracy of the approximation was determined by the comparison with the empirical distribution function, so that these graphs are only a more detailed illustration of the method. Obviously, both distribution functions and histograms are visually very close, even with the account of various peculiarities. It is clear that in each case the shape of the distribution is far from that of the Gaussian law.

Figure A10 shows the results of the Kolmogorov–Smirnov homogeneity test from the additionally simulated samples. For clarity, the standard significance levels 0.01 and 0.05 are demonstrated. The first one is exceeded by *p*-values for 84.5% samples (268 of 317), whereas the second one is exceeded in 70.7% cases (224 of 317). Thus, for the vast majority of bootstrap samples, fairly good approximation was obtained. It is also worth noting that the shape of histograms for the simulated samples is quite close to those obtained in the NASA catalog. This fact is a clear confirmation of the correctness of the approach used, since, as it has already been noted, functionally or statistically, the distance between these functions was not measured, since it was not used as a criterion.

5. Approximation of a Sample Distribution Function Based on Minimizing χ^2 Statistics

In the preceding section, a fairly high level of goodness-of-fit was demonstrated between the probability distributions obtained using the bootstrap procedure and the sifting data of the lunar regolith samples. However, this method has a number of disadvantages. Firstly, it is imperative to

interpolate points (the initial data from the NASA catalog). The method of interpolation can significantly affect the final results. Secondly, the generation of samples and their processing by the EM-algorithm takes a rather noticeable computational time that grows as the sample size increases. In this section, an alternative approach will be proposed, within which the initial grouped data are also approximated by functions that have the form of a finite log-normal mixtures, so that statistical simulation of samples is not required. This can improve the accuracy of the approximation, as well as reduce the computational complexity of data processing.

To find the parameters of the approximating function, it is required to minimize the distance between this curve and the known values (points) of the empirical distribution function. In the preceding section, it was noted that the best results were obtained for four-component mixtures, but this method can use a different value of the parameter k.

Thus, for the approximation, the function $\mathfrak{F}(x, \theta)$ of the type (13) can be used, where $\theta = \{\theta_i\}$, $\theta_i = (a_i, \sigma_i, p_i)$ and for each of the parameters the constraints mentioned above are valid (see the end of Section 3). Then, the χ^2 statistics has the form

$$\chi^{2}(\boldsymbol{\theta}) = \sum_{i=1}^{N} \frac{\left((y_{i+1} - y_{i}) - (\mathfrak{F}(x_{i+1}, \boldsymbol{\theta}) - \mathfrak{F}(x_{i}, \boldsymbol{\theta})) \right)^{2}}{\mathfrak{F}(x_{i+1}, \boldsymbol{\theta}) - \mathfrak{F}(x_{i}, \boldsymbol{\theta})},$$
(14)

where **x** and **y** are known sets of values and percentages in the total number for the corresponding particle sizes from the NASA catalog, and *n* is the number of nonzero differences $(y_{i+1} - y_i)$. It is worth noting that there are duplicate values of y_i in the original data, so it had to be taken into account during data processing. In (14), it is assumed that all y_i are different.

To find the numerical estimates of the unknown parameters θ with the account of the constraints mentioned above, we used the solution of a nonlinear programming problem by the algorithm based on the interior-point optimization [29]. The quality of the approximation was estimated by substituting the value of the statistic $\chi^2(\theta_{opt})$ with the optimal set of parameters θ_{opt} into the distribution function χ^2 with N - 1 degrees of freedom that gave the corresponding *p*-value. This procedure is described in Algorithm A2.

Examples of the application of this algorithm to real samples of the lunar regolith are shown in Figures A11–A19. The graphs show the initial data from the tables of the NASA catalog, their interpolation by the Hermite polynomials (blue solid line), and the bootstrap procedure (red line), as well as the approximating mixture obtained by the method of minimization of the statistics χ^2 (magenta line). Here, the coincidence of the curves obtained by the two methods is not necessary, since the majority of the attention is paid to the approximation of the set of initial points. The graphs show curves for the same number of components k = 4, and, for each of the methods, a different value can be used.

Figure A20 demonstrates the results of the χ^2 test for k = 4 (see formula (13)). The graphs show the standard significance levels 0.05 and 0.01. The significance level 0.05 for the χ^2 test is exceeded by the *p*-values for 92.7% samples (294 of 317) when approximated by three-component mixtures and for 93.1% of samples (295 of 317) when approximated by four-component mixtures. The significance level 0.01 is exceeded by the *p*-values in 94.6% cases (300 of 317) when approximating by three-component mixtures and for 94% samples (298 of 317) in the case of four-component mixtures. In fact, the quality of the statistical approximation is the same for both k = 3 and k = 4. This allows for using fewer parameters when solving the constrained optimization problem.

Thus, the disadvantages of the bootstrap methodology method can be overcome. The quality of the approximation in this case can be assessed as excellent. The only important point of this approach is finding the solution by conditional optimization that seriously depends on the initial approximation and can lead to incorrect results. Therefore, careful adjustment of the parameters is required, as well as the use of a series of runs to select the most successful values. The computational complexity of the proposed procedure is moderate. Therefore, multiple computations are possible.

6. Cluster Analysis of the Mixture Parameters in Approximations of the Lunar Regolith Particle Size Distributions

This section presents the results of cluster analysis of the parameters of the components of all approximating finite normal mixtures at once. In this case, for partitioning into clusters, a full set of parameters (mathematical expectations, standard deviations and weights) are used, as well as a trivial inverse transformation to go from the ϕ scale for sizes to metric (µm). The corresponding is given in Algorithm A3.

Figures A21–A24 (top graphs) demonstrate the relationship between the mathematical expectation and standard deviation for the ϕ scale as well as μ m for both methods discussed above. The size and color intensity of the points correspond to their weights (see formula (13) and the color bar on the figures). These figures essentially clarify and refine the linear approximation proposed in [30] for this dependence, which, as can be seen from Figures A21–A24, turns out to be overly rude.

The two lower plots in each of Figures A21–A24 demonstrate the partition of the parametric space into six groups by the *k*-medoids and fuzzy c-means clustering (in this case, the solution is the cluster with the maximum membership value for this element among all possible) for the bootstrap procedure and the method based on the χ^2 estimates. It is obvious that the solutions of both methods in each of the cases are quite close. It should be noted that the results of fuzzy clustering have slightly better reproducibility compared to medoids under reclustering. For the clusters obtained by both methods, weights were determined as the ratio of the number of parameters included in the given cluster to the total number of points. The results are presented in Table 1 where the rows are in decreasing order of the corresponding weights.

Bootstrap Simulations				Minimizing χ^2 Estimates			
φ Scale		μm		φ Scale		μm	
k-Medoids	c-Means	k-Medoids	c-Means	k-Medoids	c-Means	k-Medoids	c-Means
2.08%	20.19%	42.43%	42.35%	35.09%	31.94%	51.50%	41.48%
20.35%	19.95%	24.61%	25.71%	33.44%	24.45%	18.53%	28.08%
17.27%	17.90%	13.80%	13.56%	18.61%	19.09%	17.67%	20.90%
15.22%	16.25%	9.70%	9.07%	4.73%	12.22%	6.62%	5.05%
12.54%	14.51%	5.28%	5.28%	4.34%	6.23%	5.52%	4.26%
9.54%	11.20%	4.18%	4.02%	3.79%	6.07%	0.16%	0.24%

Table 1. Cluster size ratios.

The resulting clusters can be used, for example, to correlate with the chemical compounds of samples or other characteristics of the regolith.

A different representation of Figures A21–A24 is also possible, namely a three-dimensional image. In this case, the weights of the points are plotted on a separate scale, but their size when drawing is still determined by this parameter. Examples for each of the methods for metric and ϕ scales are shown in Figures A25–A28.

7. Conclusions

A version of the central limit theorem was proved for sums with a random number of independent and not necessarily identically distributed random variables in the double array limit scheme. It is demonstrated that arbitrary normal mixtures appear as the limit distribution. This result is used to substantiate the log-normal finite mixture approximations for the particle size distributions of the lunar regolith. This model is used as the theoretical background of the two different statistical procedures for processing real data based on bootstrap and minimum χ^2 estimates. It was shown that the cluster analysis of the parameters of the proposed models can be a promising tool for revealing the structure of such real data, taking into account the physico-chemical interpretation of the results. Similar methods can be successfully used for solving problems from other subject fields with grouped observations, and only some characteristic points of the empirical distribution function are given.

The statistical data processing of all samples of the lunar regolith is presented. It is based on the sequential application of the bootstrap procedure (see Algorithm A1) and the method based on minimizing the χ^2 estimates (see Algorithm A2) to each sample from the NASA catalog in order to form a set of parameters of approximating finite normal mixtures, which are further should be clustered (see Algorithm A3). The method completely described in the paper is presented in Algorithm A4.

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Appendix A. Algorithms

This section presents the above-described algorithms To reduce blank spaces on this page, we have expanded comments in the Algorithm A1 (see green marks) that have been implemented in the MATLAB programming language. For computations, the resources of a hybrid high-performance computing cluster of architecture Intel x86_64 were used: server Huawei XH 622 V3 (two processors Intel Xeon E5-2683V4 with a frequency of 2.1 GHz (16 cores), 12 GB of RAM and 2 GPU NVIDIA Tesla K80. This allowed for increasing the computing speed from 39.3–53 s, using standard desktop solutions, up to 13.9 s, obtained on the computational cluster, for one regolith sample. Thus, an almost fourfold acceleration of computations was obtained, which is especially important when implementing the bootstrap approach, for which the size of the generated samples is very important. A method based on χ^2 statistic needs less computational resources.

Algorithm A1. Simulating samples for approximation and statistical test					
1: function GENSAMPLES(ECDF, SampleSize, TestSampleSize)					
2: // r and rTest are random vectors for the inverse transform sampling					
3: // Random vectors for estimation of parameters					
4: $r \leftarrow RAND(SampleSize);$					
5: // Random vectors for Kolmogorov–Smirnov homogeneity test					
6: $rTest \leftarrow RAND(TestSampleSize);$					
7: for i=1:SampleSize do					
8: // Simulating based on the inverse transform sampling					
9: Sample(i) \leftarrow FSOLVE(ECDF, r(i));					
10: if (i≤TestSampleSize) then					
11: TestSample(i) \leftarrow FSOLVE(ECDF, rTest(i));					
12: end if					
13: end for					
14: return [Sample, TestSample];					
15: end function					

Algorithm A2. Estimation of the mixture parameters by the method of minimizing χ^2 estimates

1: **function** CHIAPPROX(**x**, **y**)

- 2: $pEmp \leftarrow DIFF(y);$
- 3: N←LENGTH(pEmp);
- 4: $\chi^2(\boldsymbol{\theta}) \leftarrow \text{STAT}(\text{pEmp}, \mathbf{x}, \mathbf{y}, \mathbf{N});$
- 5: options←CONSTRAINTS();

/ / Constraints to minimize χ^2 statistics

// Null values are excluded

// Values in ϕ scale

// Values in um

- 6: // Finding the minimum of a function (14), constrained optimization
- 7: [ChiParams, ChiPval] \leftarrow FMINCON($\mathbf{x}, \chi^2(\boldsymbol{\theta})$, N, options);
- 8: return [ChiParams, ChiPval];
- 9: end function

Algorithm A3. Clustering the parameters of the mixture approximation of the particle size distributions of the lunar regolith

1: **function** REGOLITHCLUSTERING(Params, NumClust)

- Params_{μm} ← PARAMSTOμm(Params); // Conversion of parameters from φ scale to μm
 // k-medoids clustering
- 4: $[Clust_{\phi}, Med_{\phi}] \leftarrow KMEDOIDS(Params, NumClust);$
- 5: [Clust_{μm}, Med_{μm}] \leftarrow KMEDOIDS(Params_{μm}, NumClust);
- 6: *// c-means clustering*
- 7: [FClust_{ϕ}, Centers_{ϕ}] \leftarrow FCM(Params, NumClust);
- 8: [FClus_{μm}, Centers_{μm}] \leftarrow FCM(Params_{μm}, NumClust);
- 9: Clusters \leftarrow [Clust $_{\phi}$, Med $_{\phi}$, Clust $_{\mu m}$, Med $_{\mu m}$, FClust $_{\phi}$, Centers $_{\phi}$, FClust $_{\mu m}$, Centers $_{\mu m}$];
- 10: return Clusters;
- 11: end function

Algorithm A4. Analysis of lunar regolith

1: function LUNARREGOLITH(RegolithSamples, Size ₁ , Size ₂)					
2:	INIT();	// Loading data and initializing parameters			
3:	Parpool();	// Launching tools for parallel processing			
4:	for all RegolithSamples do				
5:	// PhiSize(i) is a particle size of ith sample in ϕ sca	le			
6:	// Values(i) is a proportion of particles with PhiSiz	e(i) size			
7:	$ECDF \leftarrow FIT(PhiSize(i), Values(i));$	/ / Interpolation of ECDF			
8:	// Simulating samples (see Algorithm A1)				
9:	[Sample, TestSample] \leftarrow GENSAMPLES(ECDF, Si	ze ₁ , Size ₂);			
10:	/ / EM algorithm for finite normal mixtures				
11:	Params(i)←NORMALAPPROX(Sample);				
12:	// Approximation errors (one sample KS test)				
13:	KSERROR(Params(i), ECDF, TestSample);				
14:	// Minimizing χ^2 statistics (see Algorithm A2)				
15:	[CHIPARAM, CHIPVAL]=CHIAPPROX(PhiSize(i	i), Values(i));			
16:	end for				
17:	// Parameters clustering (see Algorithm A3)				
18:	$Clusters \leftarrow REGOLITHCLUSTERING(Params);$				
19:	PLOT(RegolithSamples, Params, Clusters);	// Visualization			
20:	return ;				
21: end function					

Appendix B. Figures

This section presents the above-described figures.



Figure A3. Soil 14259,52 ("Apollo 14").





1

0.9

Data ECDF 16 of 24



Soil 63321,9





































Figure A21. Clustering the mixture parameters (bootstrap, ϕ scale).



Figure A22. Clustering the mixture parameters (bootstrap, μm).



Figure A23. Clustering the mixture parameters (minimizing χ^2 estimates, ϕ scale).



Figure A24. Clustering the mixture parameters (minimizing χ^2 estimates, μ m).



Figure A25. Clustering the mixture parameters (bootstrap, ϕ scale), 3D.



Parameters of Mixtures (χ^2 , ϕ -scale)

Figure A26. Clustering the mixture parameters (minimizing χ^2 estimates, ϕ scale), 3D.



Figure A27. Clustering the mixture parameters (bootstrap, μm), 3D.



Figure A28. Clustering the mixture parameters (minimizing χ^2 estimates, μ m), 3D.

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