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Application of Zipf's law to estimate undiscovered Gold endowment in the Quadrilátero Ferrífero Province, Brazil

Iago Sousa Lima Costa[1], Guilherme Ferreira da Silva[1], Marcos Vinicius Ferreira[1],

[1]Geological Survey of Brazil - CPRM, SBN, Quadra 2, Bloco H, 2º Andar, Distrito Federal, Brasília-DF, Brazil, CEP: 70040-904

Abstract

Ore-forming processes act as self-organizing critical systems. These systems exist in an unbalanced nature, such that energy-release can occur as a scale-invariant power-law behavior when a certain threshold is exceeded. In ore-forming systems, the energy released from multiple transient pulses of over-pressured fluid appears to follow a power-law, since the distribution of mineral deposits in mature provinces have been shown to obey Zipf's law. Zipf's law represents a statistical relationship between the size and rank of a discrete phenomenon. In this work, we present the application of Zipf's law in estimating the undiscovered gold endowment of the Quadrilátero Ferrífero Province in Brazil. In addition, we conducted several statistical tests to validate the application of Zipf's law and discussed its limitations regarding smaller deposits. The Kolmogorov-Smirnov's minimum distance was presented as an alternative to defining the lower boundary of the Zipf's law's domain, rather than using an economic cut-off. Our results estimate the maturity of the Quadrilátero Ferrífero Province to be 65%, with potential for the discovery of at least 749 t of gold, comprising 28 deposits larger than 8 t (totaling 519 t), and 2 deposits larger than 40 t (totaling 122 t).

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*Corresponding author
Iago Sousa Lima Costa
E-mail address:
iago.costa@cprm.gov.br

1. Introduction

The modern mineral systems concept comprises all geological factors that combine to form an ore deposit, including assembly, transportation, and ore accumulation (Wyborn et al. 1994). Even when these factors are well understood, ore-forming systems occur in unpredictable, complex patterns, focused on maximizing entropy, i.e., ore-formation represents a self-organized critical system (Hronsky 2011). In such systems, energy release follows a scale-invariant power-law behavior, with spatial fractal geometry (Bak 1996). This behavior can be seen in mature mineral provinces, where the distribution of mineral deposits follows a power-law distribution when deposits are ranked in order of decreasing size (Hronsky and Groves 2008).

A power-law distribution is not an exclusive behavior of self-organized critical systems. Similar distributions are present where a free and natural competition mechanism exists, such as economics, finance, social sciences and physics (Buchanan 2008; Haldar 2018; Newman 2005). Since these events, ranked by their frequency in descending order, follow a power-law, we can also say that they obey Zipf's law (Zipf 1949).

Zipf's law has been widely employed to estimate residual mineral endowment of provinces worldwide (Davies et al. 2018; Guj et al. 2011; Lisitsin 2016; Mamuse and Guj 2011; Paliwal et al. 1986; Yigit 2011). Likewise, the results of Zipf's law assessments have been shown to be comparable to the Three-Part Assessment (Davies et al. 2019; Lisitsin et al. 2010, 2014), developed by the United States Geological Survey (Singer 1993; Singer and Menzie 2010) and widely employed by government organizations.

However, understanding mineral deposit distributions through Zipf's law is non-trivial. Specific considerations regarding the nature of the power-law need to be carefully investigated prior to assessment (Alstott et al. 2014; Clauset et al. 2009). In this work, we discuss some premises and limitations of Zipf's law, as applied to estimate the residual gold endowment of Quadrilátero Ferrífero Province.

2. Quadrilátero Ferrífero Province

The Quadrilátero Ferrífero (Figure 1) is an important metallogenic province located in the southernmost portion of the

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San Francisco Craton, in eastern Brazil. The region has been an important mining site since the early eighteenth century and, at that time, was responsible for the Brazilian colony being one of the largest gold producers in the world (Farina et al. 2016).

The region is composed of Archean and Paleoproterozoic complexes, as supracrustal sequences forming a dome-and-keel architecture, in which Paleoproterozoic belts of low metamorphic grade surround, and overlay, medium-to-high grade Archean complexes (Farina et al. 2016; Marshak et al. 1997). The metavolcanic and metasedimentary rocks of the region are grouped in two main supergroups: Rio das Velhas, of Archean age (Baltazar and Zucchetti 2007), and Minas, mainly of Paleoproterozoic age (Babinski et al. 1995).

The Rio das Velhas Supergroup represents rocks from a typical Archean greenstone belt, with the association of mafic and ultramafic, felsic volcanic and volcanoclastic rocks, and immature clastic sediments (Dorr 1969). According to Farina et al. (2016), these rocks are metamorphosed at greenschist to lower amphibolite-facies and are commonly affected by hydrothermal alteration. The Rio das Velhas Supergroup hosts the majority of the largest gold deposits in the Quadrilátero Ferrífero Province (Figure 1). These deposits are generally associated with quartz veins and/or some stratigraphic control.

According to Alkmim and Martins-Neto (2012), the Minas Supergroup can be divided into the Basal Sequence, associated with the development of a passive margin basin, and the Uppermost Sequence, consisting of submarine fan deposits related to the inversion of the passive margin. The Minas Supergroup hosts a number of gold deposits near the Ouro Preto city, on the southern portion of the Province, but the main mineral occurrences known within this unit are the Lake Superior-type iron and manganese deposits of the Cauê Formation (Farina et al. 2016).

Morro Velho is the largest and one of the oldest known deposits in the Province. According to Vial et al. (2007), gold was first discovered in the year 1700, and mining operations have occurred continuously between 1834 and 2003. Morro Velho is assumed to be an orogenic gold deposit, associated with two main mineralization styles; high grade massive sulfide-rich orebodies and low grade dark gray quartz veins with free gold (see Vial et al. 2007 and references therein). Both styles are concentrated along the interface of the carbonatic and micaceous Lapa Seca layer (Lobato et al. 2001), a hydrothermal alteration product that may have acted as a stratigraphic trap for the mineralization process (Vial et al. 2007). The most characteristic feature of mineralization at the Morro Velho deposit is its remarkable continuity, down to a vertical depth of 2450 m and extending 4.8 km down the plunge of a fold axis (Vial et al. 2007).

2.1 Deposit database

The database used for this study was first published by Lobato et al. (2016). It was produced through the compilation

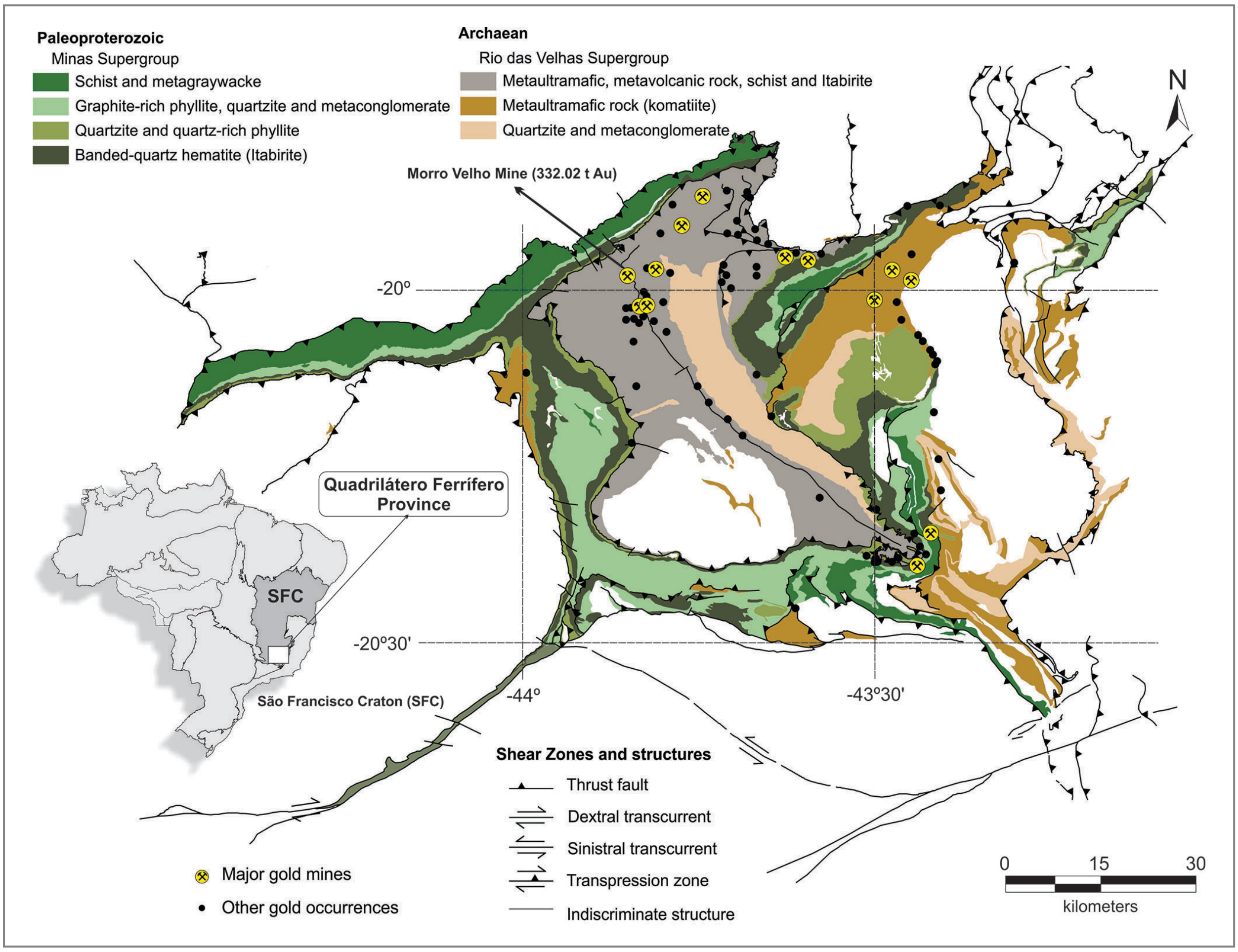

FIGURE 1 – Simplified geological map of the Quadrilátero Ferrífero Province, with emphasis on the Rio das Velhas Supergroup and the Minas Supergroup. Yellow filled circles represent the mines listed in this work (see Table 1). The black dots represent the other known occurrences and small mine sites (modified from CPRM, CODEMIG 2014).

of several previous works and historical registries of mining production, available from the early 19th century to the present day (Lobato et al. 2016).

The database contains information on the thirteen major gold deposits in the Quadrilátero Ferrífero Province, including the period of mining activity, host rock, total gold content (in some cases, the Mineral Resource or Ore Reserves Estimates) and estimated ore grade (Table 1). The average grade of the Gongo Soco deposit was missing in the original database, and so the value was obtained from Andrade and Sá (1990).

Morro Velho, Cuiabá, and Córrego do Sítio are considered world-class deposits (over 5 Moz), hosted in BIF like rocks or in quartz veins, similar to that of the Córrego do Sítio deposit. The database compiled by Lobato et al. (2016) also contains four small- (up to 10 t Au) and six medium-sized (10 - 70 t Au) deposits (Figure 2).

Even though the Quadrilátero Ferrífero is a province with a history of exploration dating back to the 17th century, new deposits continue to be discovered even in recent times. The data listed in Table 1 includes three mine sites that began operations in the early 19th century, four deposits discovered in the middle of the first half of the 20th century, five deposits discovered at the end of the 20th century and one deposit, containing almost 40 t of contained gold, that was discovered in the last 20 years.

Three deposits (Passagem de Mariana, Gongo Soco, and Maquiné) belong to a later period of the geological evolution of the Quadrilátero Ferrífero and are hosted in the Paleoproterozoic Minas Supergroup (Chemale et al. 1994), although they are deemed constituents of the same metallogenic province. According to Rudenno (1981), even genetically different types of deposits of the same commodity can be combined within a Zipf's curve. Therefore, we decided to retain these deposits in our database regarding the fact that they are considered to belong to the orogenic class, they are part of the same mineral province, and the age of ore formation is still uncertain.

3. Power-law distribution

Power-law distributions define many natural and socio-economic systems, such as the size of earthquakes (Gutenberg and Richter 1944; Newman 2004), diameter of moon craters (Neukum and Ivanov 1994), word use frequency (Estoup 1916), scientific citations (Price 1965), and city populations (Newman 2004). This distribution is applicable when larger events are rare, while smaller events are frequent (Yigit 2011). Mathematically, a quantity x follows a power-law, if its probability density $p(x)$ can be described as:

$$p(x) \propto x^{-\alpha} \quad (1)$$

where α is a scaling parameter. Equation 1 results in a heavy-tailed, where the probability tends to zero slowly to the right. Despite the low values, the tail contains a significant amount of probability (Alstott et al. 2014). Several empirical datasets follow a power-law, but not all fit this distribution. Despite being widely used to explain the size of mineral resources, some specific considerations need to be taken into account to characterize power-law distributed data (Clauset et al. 2009).

Although it is not necessary to confirm that data strictly follows a power-law, it is necessary to verify that a power-

TABLE 1 - Main orogenic gold deposits of the Quadrilátero Ferrífero Province and their respective ranks (modified from Lobato et al., 2016).

Mine	Activity	Gold (ton)	Grade (g/ton)	Host rock	Rank
Morro Velho	1834 -2003	332.02	7.19	"Lapa Seca" (Ankerite, Siderite and Carbonate)	1
Cuiabá	1934-1939; 1985-Today	174.80	9.62	Banded Iron Formation	2
Córrego do Sitio	1990-1998; 2002-Today	168.62	5.06	Quartz vein in carbonaceous schist/phyllite	3
Caeté (Pilar e Roça Grande)	1996-2001; 2008-Today	70.58	4.20	Banded Iron Formation and Quartz-veins	4
Raposos	1982-1999	67.11	9.50	"Lapa Seca" (Ankerite, Siderite and Carbonate	5
São Bento	1987-2007; 2014-Today	56.80	9.28	Banded Iron Formation	6
Lamego	2004-Today	38.53	4.45	Banded Iron Formation and metachert	7
Passagem de Mariana	1864-1974	35.08	8.33	Quartz vein in graphite-sericite phyllite and calcareous rock	8
Gongo Soco	1919-1998	13.26	30.00	Banded Iron Formation	9
Faria	1934-1995	8.97	7.44	Banded Iron Formation	10
Bicalho	1964-1996	8.78	7.25	Carbonate	11
Maquiné	1865-1896	5.28	14.49	Banded Iron Formation	12
Itabira	1987-1990	0.71	82.80	Quartz vein in Banded Iron Formation	13

law is the best available distribution for a dataset (Alstott et al. 2014). Therefore, it is necessary to compare a power-law with alternative distributions (e.g. exponential, stretched exponential, lognormal) using a likelihood ratio test (Clauset et al. 2009). This test calculates the logarithm R through the likelihood of the data using two distinct distributions. When R is positive, the first distribution is more compatible with the data; if R is negative, the second distribution prevails. However, R may be inconclusive depending on the standard deviation. Therefore, Clauset et al. (2009) proposed the use of the Vuong method (Vuong 1989) to estimate the statistical significance of R by *p-value*. When the *p-value* is less than 0.1, the assumption that R is correct is highly likely. If the *p-value* is greater than 0.1, the signal is not reliable and may be the consequence of oscillation.

Moreover, even if a power-law can describe a dataset, most empirical phenomena do not obey the power-law for all values. Therefore, it is essential to correctly estimate the lower bound (x_{min}) when the data starts to obey the power-law distribution. If x_{min} is too small, the scaling parameter α will be biased since the regression will try to fit non-power-law data (Clauset et al. 2009). If x_{min} is too large, useful data that obey the power-law will be removed. The optimal x_{min} can be obtained by the minimum of the Kolmogorov-Smirnov distance (D; Press et al. 1992), which denotes the maximum distance between the data cumulative distribution function $S(x)$ and its power-law $P(x)$:

$$D = \max_{x > x_{min}} |S(x) - P(x)| \quad (2)$$

Once the power-law domain is established, it is necessary to estimate the scaling parameter α correctly. Usually, α is obtained via least-squares linear regression (LS) between a probability density and quantity x in a log-log plot. However, more recent works have shown that the LS can lead to biased and incorrect α (Clauset et al. 2009; Corral and González 2019). A more appropriate approach is to adopt the Maximum Likelihood Estimator method (MLE; Pawitan 2001) to calculate the α through the equation (Clauset et al. 2009):

$$\alpha \cong 1 + n \left[\sum_{i=1}^{n} ln \frac{x_i}{x_{min} - \frac{1}{2}} \right] \quad (3)$$

where $x_i > x_{min}$, and represent the quantities in a dataset where $i = 1,...,n$.

4. Zipf's law

Zipf's law (Zipf 1949) represents a discrete version of the 80/20 Pareto theorem (Pareto 1927), which correlates size and ranked data through a power-law distribution. This statistical relationship arises when there are unobserved variables in a particular phenomenon (Aitchison et al. 2016). Since the largest event of a system is a known variable, the other events can be estimated through:

$$y_r = M_C \cdot r^{-k} \mid k > 0 \;\&\; r \geq 1 \quad (4)$$

where M_C denotes the largest event in the system, r the event rank and k the decay constant ($k = \alpha - 1$). The value of k is dependent on the nature of the system. Several works focusing on the assessment of mature provinces endowment have reported k very close to 1 (Guj et al. 2011; Lisitsin 2016; Mamuse and Guj 2011; Paliwal et al. 1986; Yigit 2011). Therefore, if we assume that Zipf's law with k = 1 can explain the endowment of an area, the total endowment (M) can be represented by the sum of the harmonic series $c + \frac{c}{2} + \frac{c}{3} ... + \frac{c}{n}$ or:

$$M = \sum_{r=1}^{n} M_C \cdot r^{-1} \quad (5)$$

where M_C is the endowment of the largest deposit, and the n (or x_{min} in power-law denotation) is the maximum rank, which can be defined through the Kolmogorov-Smirnov's distance.

The total endowment, as well as Zipf's law, is highly sensitive to the largest deposit (M_C). Usually, the largest deposits are discovered in the early stages of exploration (e.g. Morro Velho deposit), however, rough estimates of the largest deposit can significantly modify the assessment of undiscovered mineral endowments (Hronsky and Groves, 2008). Therefore, Zipf's law performs better in mature mineral provinces, where the largest deposit is likely to have been discovered and fully delineated.

5. Quadrilátero Ferrífero gold endowment assessment

To formulate the gold endowment assessment models of Quadrilátero Ferrífero, we assume that the largest deposit, Morro Velho, is known. This presumption is robust since the deposit has a substantial size, and the process of gold mining in the region began in the early-17th century (Joffily 1988).

When ranked, the Quadrilátero Ferrífero gold deposits present a negative power function. In a first attempt, the coefficient k was estimated through the LS method, which resulted in k = 1.050 with an $R^2 = 0.94$ (Figure 2a). The LS method shows a high similarity between the known gold deposits and Zipf's Law, suggesting a high degree of maturity for the province, but not yet justifying the application of Zipf's law to estimate missing endowment. However, by estimating k via MLE, the value given is; k = 0.20, i.e., the gold deposits are far from geological equilibrium (k = 1.0). This value also suggests that the gold deposits do not follow a power-law, but in-fact follow another distribution. Table 2 shows the logarithm R and *p-value* for the distribution: Exponential, Truncated Power-law, Stretched Exponential, and Lognormal positive. Here, the power-law presents a better distribution than the exponential method (R>0), but not the remaining alternative methods. However, the *p-values* for the other methods are considerably high (*p-value* ≥ 0.38), which does

TABLE 2 - Comparison between the power-law and alternative distributions for the gold deposits of the Quadrilátero Ferrífero.

Distribution Method	logarithm	*p-value*
Exponential	0.21	0.90
Truncated Power-Law	-0.38	0.38
Stretched Exponential	-0.27	0.69
Log-normal positive	-0.20	0.71

not favor any of the alternative distributions for this dataset, i.e., this result does not reject the power-law hypothesis.

The log-log-rank-size plot (Figure 2b) shows a strong correlation between the gold deposits and the Zipf's curve until rank 8, where the deposits deviate from the Zipf's curve. Furthermore, the Kolmogorov-Smirnov's minimum distance (Figure 3a) also is located at rank 8 (or x_{min}= 35.08), i.e., the current Quadrilátero Ferrífero database only obeys a power-law up to rank 8. It is also common to examine the similarity between the Complementary Cumulative Distribution Function (CCDF) of the dataset and the power-law over a log-log plot (Figure 3b). Here, the data clearly does not fit a power-law, even when k = 0.20.

Nevertheless, several authors, with robust databases, show that the distribution of mineral deposits within a province follows a power-law, specifically the Zipf's Curve with *k = 1* (Guj et al., 2011; Hronsky and Groves, 2008; Mamuse and Guj, 2011; Paliwal et al., 1986). In this case, we have two hypotheses: either the size of deposits from rank 8 have been incorrectly assessed; or there are gaps between the ranks, which can be interpreted as undiscovered deposits, deposits are known but not present in our database or misrepresented endowment of known deposits (Mamuse and Guj, 2011). The first hypothesis is unlikely to present a material difference because distinct companies, spatially distributed in different geological environments, characterized the deposits. The second hypothesis is admissible, since there is still potential for significant undiscovered mineralization in the Quadrilátero Ferrífero, despite its high maturity. Therefore, we assume the second hypothesis as valid in this study.

Given the acceptance of Zipf's law for assessing the endowment of the province, it is possible to fit the known gold deposits to the Zipf's curve and define the missing ranks (Folinsbee 1977). Table 2 shows the comparison between a power-law and alternative distributions for the dataset after ranks have been 'filled' following Zipf's law. For this new database, we obtained an acceptable logarithm R (*p-value* < 0.1) for the exponential and log-normal positive distributions, wherein both cases a power-law is recognized as the best distribution (as opposed to the inconclusive test for the initial database).

TABLE 3 - Comparison between the power-law and alternative distributions for the gold deposits of the Quadrilátero Ferrífero adjusted with Zipf's law.

Distribution Method	logarithm	*p-value*
Exponential	389.48	~ 0.00
Truncated Power-Law	-0.23	0.49
Stretched Exponential	2.33	0.32
Log-normal positive	47.63	~ 0.00

Although the filled database follows a power-law, this is unlikely to fit all deposits. For the filled database, the Kolmogorov-Smirnov's minimum distance moves from x_{min}=35.08 to x_{min}=4 (Figure 4a). Even if values were to be estimated below the smallest deposit (Itabira; 0.71 t), x_{min} converges to x_{min}=4. The x_{min} equals to 4 t is not an obstacle since smaller deposits are uneconomic and generally not reported. Using MLE, the new α for the filled deposits produces x_{min}=0.89, which is very close to the desired k = 1. A log-log plot (Figure 4b) shows the high similarity between the CCDF of Zipf's law filled deposits and the power-law regression (MLE).

The Zipf's curve adjustment (Figure 5) shows a total undiscovered endowment of 749 t, distributed into 67 deposits and small prospects, with 28 deposits large than 8 t (totaling 519 t), and 2 deposits large than 40 t (totaling 122 t). Considering the current gold endowment (approx. 1,395 t), the Quadrilátero Ferrífero Province has an estimated total gold endowment of approximately 2,144 t. The results in the province show an exploration maturity of 65% and a residual potential of 35% when considering deposits containing greater than 4 t gold.

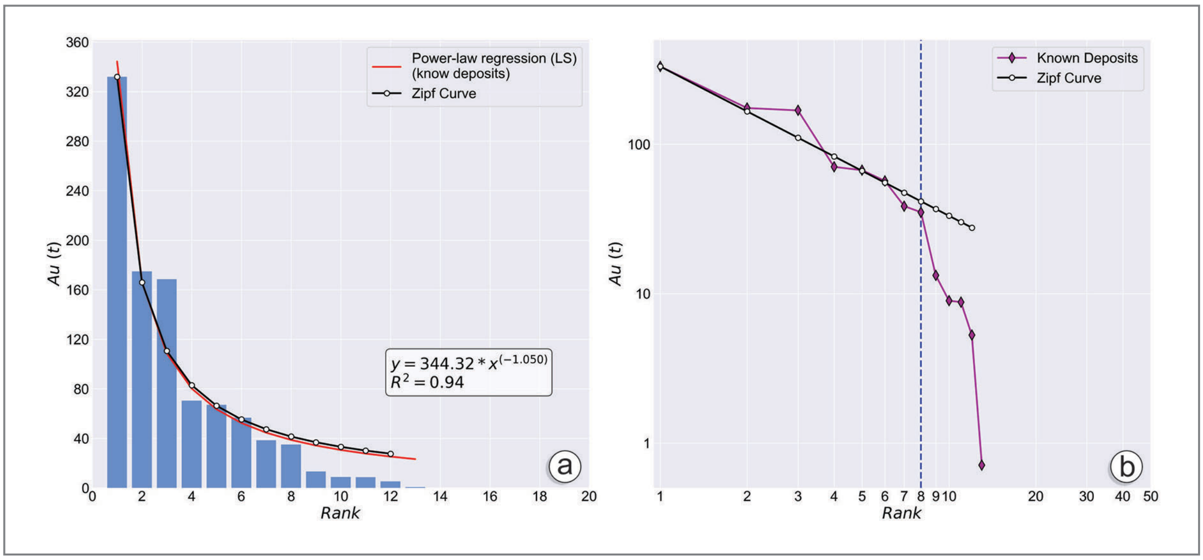

FIGURE 2 - a) Descending order rank of known gold deposits of Quadrilátero Ferrífero with the power-law curve estimated by least-square regression (red line) and the Zipf's curve (black line). b) Log-log rank-size plot showing the known gold deposits and the Zipf's curve (black). The dashed vertical blue denotes rank 8, where the deposits start to move away from the Zipf's curve.

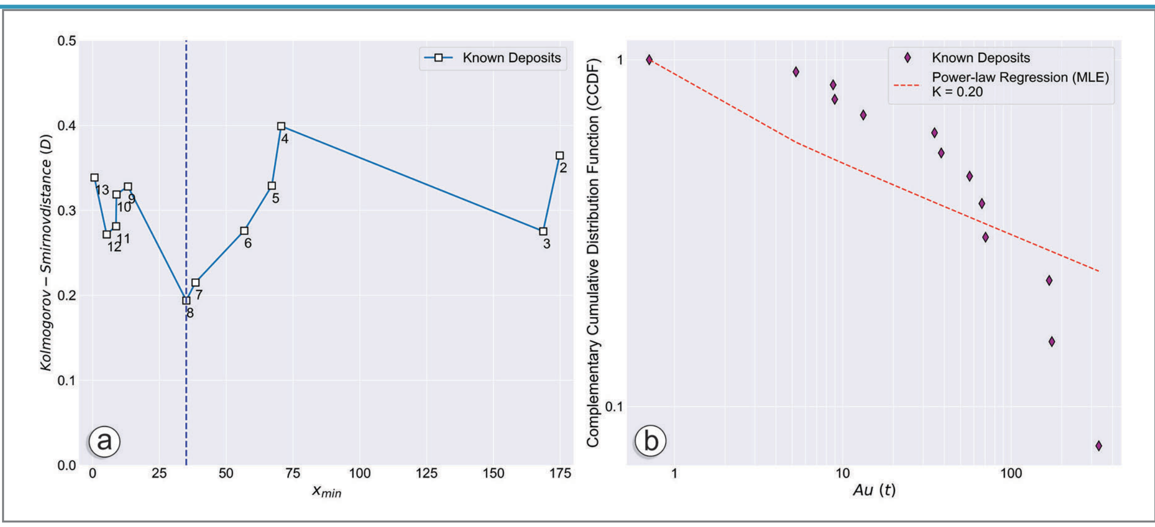

FIGURE 3 - a) Kolmogorov-Smirnov's distance for the known deposits of Quadrilátero Ferrífero. The minimum of the function (dashed vertical blue line) stands at rank 8 (or x_{min}=35.08). b) The log-log plot between the Complementary Cumulative Distribution Function (CCDF) and the gold deposits. The dashed red line shows the power-law regression using the Maximum Likelihood Estimator (MLE) with k = 0.89.

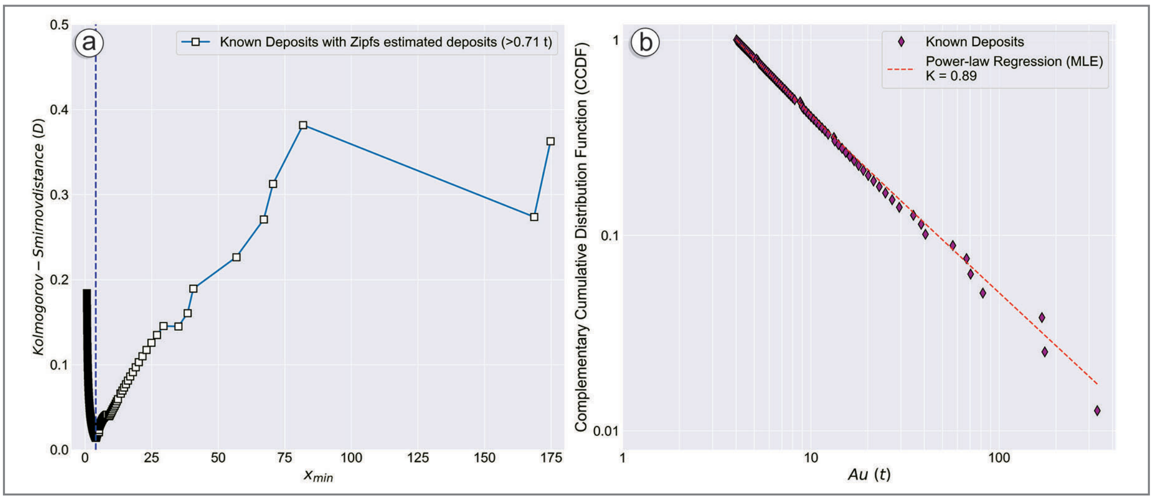

FIGURE 4 - a) Kolmogorov-Smirnov's distance for the known deposits aggregated with Zipf's estimated deposits. The minimum of the function (dashed vertical blue line) stands when x_{min}=4. b) The log-log plot between the Complementary Cumulative Distribution Function (CCDF) and the filled deposits. The dashed red line shows the power-law regression using the Maximum Likelihood Estimator (MLE) with k = 0.89.

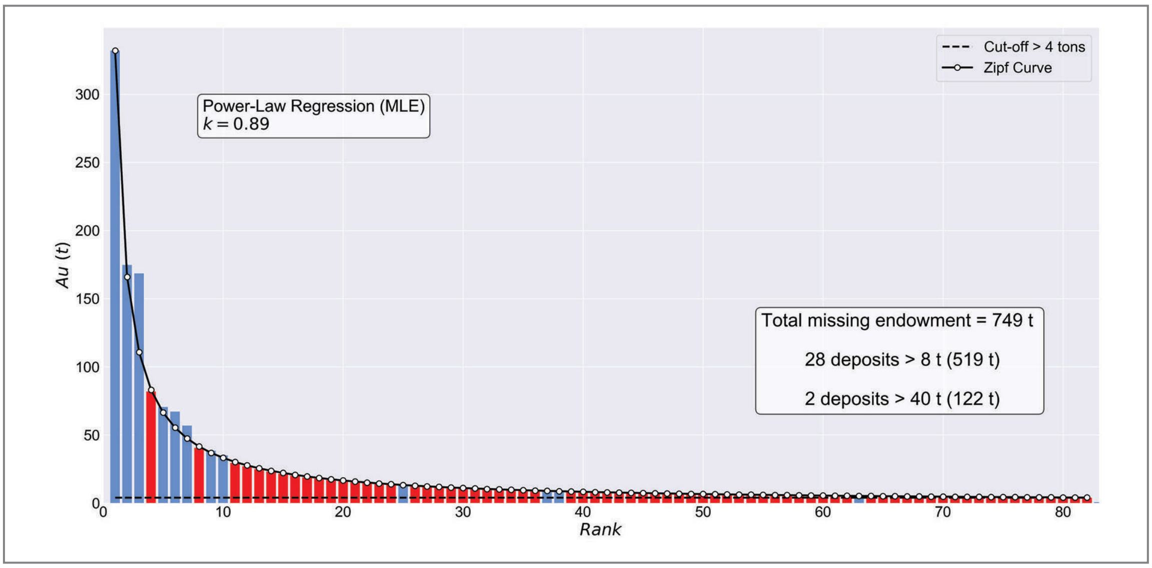

FIGURE 5 - Known gold deposits (blue bars) fitted to the Zipf's curve, and the rank gaps filled with undiscovered deposits (red bars) with an endowment greater than 4 t.

6. Discussion and conclusions

Deposits in mature provinces worldwide follow a power-law, particularly the Zipf's law (Hronsky and Groves 2008). The geological condition achieves a stable equilibrium when the Zipf's law has a decay coefficient k = 1 (Paliwal et al. 1986). Therefore, Zipf's law presents a robust statistical estimate for the mineral endowment in a province. However, statistical tests must be performed to demonstrate the Zipf's law as valid for a mineral database. In this work, we employed novel methods to evaluate the reliability of the assessment through Zipf's law, and estimate the undiscovered gold endowment in the Quadrilátero Ferrífero Province, Brazil.

We showed that the estimation of the decay coefficient k, using the least-squares method, could lead to incorrect estimates. The least-squares method measured k=1.05 for the gold database, i.e., the gold deposits in our database already follow a stable Zipf's law. However, the maximum likelihood estimator calculated k = 0.20. Therefore, if deposits actually follow Zipf's law, there is a significant number of gaps in the rank/tonnage association. These gaps can be filled until the geological condition achieves a stable equilibrium (k = 1).

However, even a dataset containing filled gold deposit ranks based on a Zipfian distribution does not follow a power-law for the entire dataset domain. The minor estimated deposits in the asymptotic tail of Zipf's curve lead to an overestimated endowment (Quirk and Ruthrauff 2006). The most common strategy is to employ an economic cutoff. In some cases, the economic cutoff may represent a value in the Zipf's tail that does not respect a power-law; therefore, the Kolmogorov-Smirnov's distance represents an excellent approach to establish the mineral endowment domain. The Kolmogorov-Smirnov's test showed that the filled gold deposits database only follows a power-law down to 4 t. In other words, gold endowment estimates below 4 t could lead to inaccurate estimates.

The application of Zipf's law showed potential for 749 t of undiscovered endowment that comprises 28 deposits large than 8 t (totaling 519 t), and 2 deposits large than 40 t (totaling 122 t). The current and estimated gold deposits show a high correlation with a power-law distribution through the likelihood ratio test. The fitted power-law regression shows a coefficient close to 1; k=0.89. This Zipf's estimation is highly dependent on the largest deposit endowment, and any variation can induce significant changes in the province's total endowment. For example, increasing the Morro Velho endowment from 332.03 to 342.03 t would increase the total undiscovered endowment from 749 to 785 t. In other words, the estimated total endowment represents a current scenario (e.g. Guj et al. 2011), which may be altered with the discovery of new deposits or re-evaluation of known deposits.

Additionally, some undiscovered endowment may represent missing known deposits or uncatalogued historical production. For example, Joffily (1988) estimated the gold withdrawal of at least 127 t during the Brazilian colonial period (1500 - 1808). Moreover, the Zipf's law method employed in this work does not regard the grade of ore in each deposit, i.e., it is possible to estimate the amount of gold remaining to be discovered, but this mineralization may not necessarily be economically viable to extract.

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