Aboav-Weaire's and Lewis' Laws—A Review

S. N. Chiu*

Freiberg University of Mining and Technology, TU Bergakademie Freiberg, Institut für Stochastik, D-09596 Freiberg, Germany

A brief review of Aboav's law (the relationship between the average number of edges of a randomly chosen neighboring cell of a typical n-cell and n) and Lewis' law (the relationship between the mean area of a typical n-cell and n) is presented. These two laws now play central roles in Rivier's maximum entropy theory of statistical crystallography. A discussion of the mathematical forms of these laws is our emphasis in this article.

INTRODUCTION

Froths (so called by physicists) or random tessellations (so called by mathematicians), which form an important class of space-filling cellular structures, have been studied extensively, both analytically and empirically. Many first-order characteristics were found (Mecke [1], Møller [2], Okabe et al. [3], Rivier [4], and Stoyan et al. [5]. However, analytic and empirical results for conditional characteristics appear not so frequently in published data. Exceptions are empirical studies of m_n (the average number of edges of a randomly chosen neighboring cell of the typical cell of a planar tessellation under the condition that it has n edges; such a cell is called the "typical n-cell") and of A_n (the mean area of the typical n-cell of a planar tessellation). But these results are widely scattered in biological, geographical, mathematical, and physical literature. Scientists, mainly physicists, have struggled for a long time to find the relationship between n and m_n (Aboav's law) or A_n (Lewis' law). Aboav's law or Aboav-Weaire's law, which was first investigated empirically by Aboav [6] with the original aim of under-

standing the mechanism of the growth of polycrystals, and Lewis' Law, which was found originally in epidermal studies of *Cucumis* (Lewis [7, 8]), are now observed in many naturally cellular networks, even in the cellular structure of the administrative divisions of a country (Le Caër and Delannay [9]) or in some similar networks (Boots [10] and Pignol et al. [11]). These two laws now play central roles in Rivier's [4] maximum entropy theory of statistical crystallography.

Here, the studies of Aboav's law and Lewis' law is briefly reviewed. The ultimate aims are (a) to give a review of the development of the theory and a collection of important results, and (b) to consider these problems in the frame of stochastic geometry to see the possibility of further theoretical studies. Therefore, we emphasize the mathematical forms of the laws. A proof of an exact identity related to Aboav's law, using techniques in stochastic geometry, is given in Appendix A.

Other texts on the subject can be found in Biarez and Gourvès [12], Bideau and Dodds [13], Bideau and Hansen [14], Dormer [15], Getis and Boots [16], Gibson and Ashby [17], Gorden [18], Guinier [19],

Okabe et al. [3], Smoljaninov [20], Stoyan et al. [5], Stoyan and Stoyan [21], Thompson [22], and Weaire and Rivier [23].

Physicists have not always stated explicitly their underlying models, except Rivier [24, 4] who stated that all cells should be convex polygons in the two-dimensional (2D) space (polytopes in the d-dimensional space), and some of the arguments are not true in general when nonconvex polygons are allowed. Thus, we assume in the following context that all polygons are convex. Moreover, we assume all tessellations are stationary and in *ordinary equilibrium state*, that is, there are always three edges emanating from each node in a planar tessellation, which means all vertices have the same coordination number 3, where the networks (froths) are of trivalent vertices.

As this is a review article, I prefer to write down all the authors of published data; so even if an article is of seven authors, the abbreviation et al. is not used. Appendices A and B contain some relevant excerpts of two not-yet published works.

ABOAV-WEAIRE'S LAW

Lewis [25] observed that in planar random tessellations seen in epithelia, the few-edged cells have a remarkable tendency to be in contact with many-edged cells and vice versa.

Aboav [6] found empirically that

$$m_n = 5 + \frac{8}{n}, \qquad (1)$$

and in his example, the average experimental error was less than 3%. This formula is an approximation; and because of the following relation, the constants 5 and 8 cannot be true in general.

Denote $\mu_2 = Var(N)$, $E(N^2) = \mu_2 + E(N)^2$, where N is the random number of the typical cell edges, with the distribution mass function $\{p_n\}$. As we have assumed that the tessellation is stationary and the number of edges emanating from each vertex of a cell is always 3, then $E(N) = 6$. The following rule, called *Weaire's sum rule* (Weaire [26]), was used frequently in published data,

$$\Sigma m_n n p_n = \mu_2 + 36 = E(N^2). \qquad (2)$$

The leftmost term of Eqn. (2) is the mean total number of the edges of all neighboring cells of a typical cell. Weaire's proof is rather "heuristic." (Every cell of n edges will be counted n times. However, we have to perform this counting process among infinitely many cells! Then, we have to divide the total number of edges of the neighboring cells of all (infinitely many) cells by the total number of (infinitely many) cells. Fortunately, a rigorous proof, using Mecke's [1] Palm method, can be constructed easily, and is given in Appendix A (see also Chiu [27] and Weiss [28]). Lambert and Weaire [29] have given a generalization of this sum rule, but this generalization did not provide a new insight of Aboav's law and was shown (Le Caër and Delannay [30]) to be a simple consequence of the symmetry of the topological correlation functions $A_{nj} = A_{jn}$, where $A_{jn} = M_j(n)/p_j$, and $M_j(n)$ is the mean number of j-edged neighboring cells of a typical n-cell (see the sections about Rivier's microreversible system and maximum entropy method and about Le Caër's topological lattice model).

Weaire [26] used the sum rule (2) and proposed

$$m_n = 5 + \frac{6 + \mu_2}{n}, \qquad (3)$$

and Aboav [31] suggested that

$$m_n = (B_1 + B_2\mu_2) + \frac{B_3 + B_4\mu_2}{n},$$

where the B_i's are constants for $i = 1, 2, 3$, and 4. By Weaire's sum rule, it becomes

$$m_n = 6 - a + \frac{b\mu_2}{6} + \frac{6a + (1 - b)\mu_2}{n}, \qquad (4)$$

where a and b are constants. Now (3) is a special case of (4) with $a = 1$ and $b = 0$, and (1) is a special case of (3) with $\mu_2 = 2$. Experimental studies suggested that $b = 0$, and $a = 1.2$, with average error less than 2%, but the deviations showed signs of being systematic (Aboav [33]). (4) with $b = 0$ can be written as

$$m_n = 6 - a + \frac{6a + \mu_2}{n}, \qquad (5)$$

and can also be expressed as

$$\frac{m_n n - E(m_N N)}{n - E(N)} = 6 - a. \tag{6}$$

Aboav [31] raised two questions: (a) is $a = 1.2$ a purely geometrical property or does it depend on a physical property of the system; (b) is it directionally dependent?

An artificial example showed that the linear law

$$m_n = B_1 + \frac{B_2}{n}$$

does not hold (Aboav [31b]). On the other hand, $a = 1.2$ was found to be valid for three different natural physical systems (Aboav [32]). In natural structures, deviations from Aboav's law mainly occur outside the range $5 \leqslant n \leqslant 8$.

Boots [34] considered Voronoi polygons and showed that a can be less than 0.6 and can depend on the aggregation of the polygons' generating points. That is to say, a varies with different systems. Analytic results for a special model in Le Caër and Delannay [30], simulations of Delannay and Le Caër [35], Fortes and Andrade [36], and Peshkin, Strandburg, and Rivier [37] and experiments of Gervois, Troadec, and Lemaître [38], Lemaître, Gervois, Bideau, Troadec, and Ammi [39], and Lemaître, Gervois, Troadec, Rivier, Ammi, Oger, and Bideau [40] corroborated the variability of a.

Experiments showed that there is a relationship between the number of edges of a typical cell and its size. Therefore, unlike $E(N) = 6$, μ_2 is not only a topological quantity but also depends on metric consideration.

Aboav [32] conjectured that a is a metrical parameter whose value for the systems considered is determined by a geometrical condition governing the angle at which the cell edges meet. In his later work [41], he introduced α, the differential longitudinal dispersion, and renamed it in [33] as differential metrical dispersion, by

$$\alpha = \frac{S_X - S_Y}{S_X + S_Y}. \tag{7}$$

In Aboav's notation, X is the typical edge length of the cells of a tessellation; Y is that of its dual. A dual (not uniquely defined to a tessellation) is constructed in the following way. For each cell in the tessellation under investigation, choose its centroid or generating point (if existing) as a dual node. Whenever two cells are neighboring to each other, the corresponding nodes in the dual are joined together by adding an edge to them. So a Delaunay tessellation is a dual of a Voronoi tessellation, but we can also choose the centroids of a Voronoi tessellation to produce a dual. And

$$S_X = \frac{Var(X)}{E(X)}, \qquad S_Y = \frac{Var(Y)}{E(Y)}.$$

Therefore, α is a geometrical parameter only.

The results from six natural systems (used centroids to produce duals) and four Voronoi tessellations (used generating points to produce duals) showed empirically that

$$a = 6\alpha, \tag{8}$$

with the range that $\alpha \in [0.08, 0.2]$ and $\mu_2 \in [0.6, 2.9]$.

Aboav's [31] experimental results suggested that α and μ_2 are independent but that S_X, S_Y and μ_2 are strongly correlated in some cases and are less correlated in some other cases. With $a = 6\alpha$, (5) can be reexpressed as

$$m_n = 6(1 - \alpha) + \frac{36\alpha + \mu_2}{n}, \tag{9}$$

for $\alpha \in [0.08, 0.2]$ and $\mu_2 \in [0.6, 2.9]$. Aboav [33] used different methods to produce generating points of Voronoi polygons, and the results showed no violation of the equation.

Delannay and Le Caër [35], Fortes and Andrade [36], Le Caër and Delannay [30], and Pignol, Delannay, and Le Caër [11] have observed that all tessellations considered so far were rather similar in their geometrical aspect in that the cells were fairly regular polygons and μ_2 was small, usually less than 2, and never exceeds 5 in natural structures. This may be the reason why a in natural froths is always close to 1. Le Caër and Delannay [30] proposed that a decreases when μ_2 increases and becomes negative for large μ_2. Although this suggestion differs from Aboav's [31], it has been empirically justified; see also Godrèche, Kostov, and Yekutieli [42] ($\mu_2 = 10.5$ and $a \approx -1$)

and Peshkin, Strandburg, and Rivier [37] (μ_2 = 12.69 and a = −1.33).

Fortes and Andrade's [36] model is not only of large μ_2 but also looks different from many natural structures. They have constructed two kinds of networks of Poisson-distributed straight lines. One was simply the tessellation formed by a Poisson line process on the plane. They called such a tessellation a 4-regular network (as there are always 4 edges emanating from each vertex). The other kind was generated similarly, by drawing successive random straight lines on the plane, but a new line was interrupted at the points of intersection with previously drawn lines, forming T-junctions. These tessellations were called 3-regular networks because there are always three edges emanating from each vertex. These tessellations looked rather different from Voronoi tessellations and those considered by other physicists. By studying a 3-regular network with μ_2 = 9.11, they claimed that m_n in that network was still well presented by Aboav's law for a = 0.35 despite some deviation for large n. They thought that this deviation was due to their *poor statistics* but without further comment. This study led to the conclusion that Aboav's law may have a great general applicability to random networks. These authors observed that the linear relation is not more than a good approximation, and the exact determination of m_n may require the knowledge of the probability that an edge belongs to two cells of n- and j-edges. (We will discuss this in the next section.)

Deviations from the law, however, appear in Poisson Voronoi tessellations (Boots [43]). To overcome this, Aboav [44] proposed

$$m_n = E(N) + \frac{2}{n} + \frac{E(N^{1/2}) - n^{1/2}}{2}, \qquad (10)$$

but the drawbacks (suggested by Le Caër and Ho [45] are (a) it departs too much from the basic linear law, and (b) it is also not valid for all tessellations. Weaire's sum rule (2) requires that

$$\mu_2 = 2 + \frac{E(6N^{1/2}) - E(N^{3/2})}{2}. \qquad (11)$$

This is approximately true for the Poisson Voronoi case but not for Voronoi polygons generated by eigenvalues of random matrices (Le Caër and Ho [45]).

Boots and Murdoch [46] suggested

$$m_n = B_1 + \frac{B_2}{n} + \frac{B_3}{n^2}, \qquad (12)$$

with B_1 = 5.01, B_2 = 9.68, and B_3 = −11.18 for Poisson Voronoi polygons. And Weaire's sum rule (2) requires

$$m_n = 6 - a + \frac{b}{6}E\left(\frac{1}{N}\right) + \frac{6a + \mu_2}{n} - \frac{b}{n^2}. \qquad (13)$$

Le Caër and Ho [45] had another suggestion,

$$m_n = B_1 + \frac{B_2}{n} + B_3 n, \qquad (14)$$

which yields, using Weaire's sum rule (2),

$$m_n = 6 - a + b\left(6 + \frac{\mu_2}{6}\right) + \frac{6a + \mu_2}{n} - bn. \qquad (15)$$

A fit of m_n values for Poisson Voronoi tessellation gives a = 0.2332 and b = 0.06343, but (15) is not valid for all n as m_n < 0 for $n \cong$ 195 (although the probability of $\{N$ = 50$\}$ is already of order 10^{-75}; see Drouffe and Itzykson [47]).

They also claimed that the ratio b/a is related to the *ageing* of their physical structures that changes the cell shape correlations. The *equilibrated* cells may correspond to b/a = 0. The Poisson Voronoi tessellation gives a large b/a ratio and so it is a *young* structure (see also Rivier [48]).

In addition, they studied the conditional second moment of the number of edges of a typical n-cell randomly chosen neighbor, $\sigma_m^2(n)$, and found that empirically

$$\sigma_m(n) = \beta(m_n - \gamma) \qquad (16)$$

with β = 0.241 and γ = 4.723 in Poisson Voronoi cells. Using sampling theory, they obtained that the estimator of the standard derivation (s) of m_n is

$$s(m_n) = \frac{\sigma_m(n)}{\sqrt{ICp_n}}, \qquad (17)$$

where I is the number of simulations, with C cells and p_n the probability that the typical cell has n edges.

Lauritsen, Moukarzel, and Herrmann [49] studied dynamic Voronoi diagrams and also found deviations of Aboav's law.

EXACT IDENTITY

Actually, it can be shown by using Mecke's [1] Palm method (Chiu [27]) or by some simple but not mathematically rigorous geometric arguments (Chiu [50]), that for $p_n > 0$,

$$m_n = 5 + \frac{6}{n} + \frac{cov[k(n,N),N]}{np_n},\qquad(18)$$

where $k(j,n)$ is the mean number of j-edged cells belonging to the *complex* or the *system*, which is the union of the typical cell and its neighboring cells (this *complex* is the main object of this study), under the condition that the typical cell has n edges when $p_n > 0$ and is zero otherwise. The proof taken from Chiu [27] is given in Appendix A.

This covariance is not yet known but should depend on p_n. The mean value $k(j,n)$ is not new to physicists. Delannay, Le Caër, and Khatun [51], Le Caër and Delannay [9, 30], Mombach, de Almeida, and Iglesias [52], Peshkin, Strandburg, and Rivier [37], and Pignol, Delannay, and Le Caër [11] have studied $M_j(n)$ (the average number of j-edged neighboring cells of a typical n-cell). $k(j,n)$ and $M_j(n)$ can be connected by a simple relation,

$$k(j,n) = M_j(n) + \delta_{nj},\qquad(19)$$

where

$$\delta_{nj} = \begin{cases} 1 \text{ when } n = j \\ 0 \text{ when } n \neq j. \end{cases}\qquad(20)$$

Simulation and experimental data given by physicists (see Appendix A) showed that the behavior of that covariance (as a function of n) is rather similar to that of p_n, when p_n increases (decreases), the covari-

ance increases (decreases). Unfortunately, the ratios between the covariances and p_n's do not show a special pattern (Chiu [27]). Nevertheless, Eqn. (18) can still provide some insight for the linear law, especially that the first two terms are so close to (1) and (3) and that "magic" number[1] "5" appears again (see Peshkin, Strandburg, and Rivier [37] for a discussion of this "magic" number). It is suggested that perhaps physicists should investigate $k(j,n)/p_j$ more deeply.

Moreover, as first observed by Fortes and Andrade [36], natural tessellations studied so far were usually with fairly regular cells and low variances of the number of edges of a typical cell. This means that there were only a few data points; so, it is not surprising that a linear regression line provides a good fit of the m_n values, especially when the exact relation already contains a linear part.

RIVIER'S MICROREVERSIBLE SYSTEM AND MAXIMUM ENTROPY METHOD

Rivier [48] considered a microreversible system—a system where elementary topological transformations (neighbor switching, T_1 [Fig. 1]; face disappearance, T_2 [Fig. 2], and their inverses, and mitosis, or cell division, which is an iterated inverse T_2 transformation) can occur (when possible, e.g., T_2 is not possible in Voronoi tessellations where the number of cells is conserved) independently of each other in space or time, without affecting the statistical equilibrium
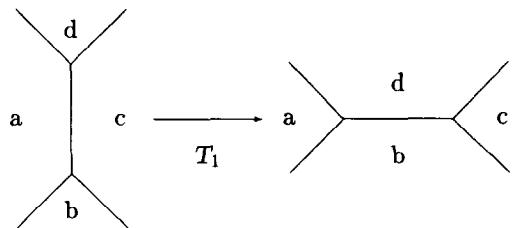

FIG. 1. Neighbor switching, T_1 (after Rivier [48]).

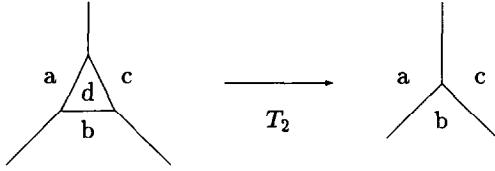[1] The term "magic number" was first used by Le Caër and Delannay [30].

FIG. 2. Face disappearance, T_2 (after Rivier [48]).

of the structure. Discussions of these transformations can be found in Rivier [24, 4].

Recursive formulas for both T_1 and T_2 have been obtained in Rivier [48]; and with the assumption that there is no dependency beyond nearest neighbors in T_1, the solution (for both T_1 and T_2) was

$$m_n = 5 + \frac{6 + \mu_2}{n}. \qquad (21)$$

However, the argument for T_1 was shown to be incorrect by Peshkin, Strandburg, and Rivier [37]. The solution for T_1 was found to be

$$m_n = 5 + \frac{\mu_2}{6} + \frac{6}{n},$$

whereas (21) is still valid for T_2. These authors argued that if the ratio of T_1 to T_2 were independent of n, Aboav's law would still be obtained from this microreversibility with the constant term intermediate between 5 and $5 + \mu_2/6$. But they also believed that it is not a realistic assumption. They simulated these two elementary topological transformations and the results agreed with their predictions.

As this microreversibility cannot be used to derive Aboav's law, they used the maximum entropy method developed by Rivier [4, 24, 48, 53–55] and Rivier and Lissowski [56] to show that the entropy $S \equiv -\Sigma_n p_n \ln(p_n/q_n)$, where q_n is a *prior* of p_n (see Rivier [4]), will be maximized if

$$M_j(n) = A_j + B_j n, \qquad (22)$$

for some A_j and B_j depending on j only, and so

$$nm_n = \sum_j jM_j(n) = \left(\sum_j jA_j\right) + \left(\sum_j jB_j\right)n \qquad (23)$$

(this form is called a *linear* Aboav's law). However, not only their own simulation but

also those of Delannay, Le Caër, and Khatun [51], Le Caër and Delannay [9, 30], and Mombach, de Almeida, and Iglesias [52] showed empirically that $M_j(n)$ departs from the linearity. When they studied Lewis' law Gervois, Troadec, and Lemaître [38] suggested, that $\Sigma_n(1/n)p_n$ may be fixed. Le Caër and Delannay [30] also discussed some probably missing topological constraints such as the positivity of $M_j(n)$, the impossibility of two 3-edged cells neighboring each other $[M_3(3) = 0]$, the correlation between the number of 3-cells and 4-cells, and the triplet correlations.

Chiu [50] (see Appendix B for some excerpts) showed that (22) is only a *sufficient but not necessary condition* for S to be maximized under those constraints considered in Peshkin, Strandburg, and Rivier [37]; that is, although the linear form (22) implies that S is maximized, when S is maximized, it is not necessarily that (22) holds. Actually if Peshkin, Strandburg, and Rivier's [37] arguments were applied on $k(j,n)$, we would conclude that $k(j,n)$ is also linear in n. It is impossible to have both $M_j(n)$ and $k(j,n)$ being linear in n. On the other hand, we can, by using the same arguments, obtain a nonlinear Aboav's law with the same value of S. Chiu [50] concluded that Rivier's maximum entropy method cannot be used to derive or prove Aboav's law, and the equivalence between the statistical equilibrium and the maximum value of S is in doubt.

Using a microreversibility argument similar to that in the planar case, Rivier [48] proposed a 3D version of Aboav's law for $m_f(n)$, the average number of edges of the neighboring faces of an n-edged face of a typical cell, under the condition that it has f faces. But this relation has not yet been checked against simulation or experimental data. Moreover, parts of this work are incorrect because the author has not distinguished a typical face and a face of a typical cell, which have different distributions. Perhaps, it is more interesting to investigate the mean number of edges of the neighboring faces of a typical n-edged face or the mean number of faces of the neighboring cells of a typical f-faceted cell in 3D.

THREE-DIMENSIONAL ABOAV'S LAW

Aboav [57, 58] studied a 3D metal structure and obtained a 3D Aboav's law for the mean number of edges of the neighboring faces of a typical n-edged face. (Unlike soap foam, metal is opaque; so, it is difficult to investigate its topological properties in 3D). Observing *directly* the shape of individual grains of pure aluminum separated from a polycrystal, he found that after grain growth,

$$m_n^{(3)} = E(N) + \frac{\mu_2}{n}, \qquad (24)$$

where $m_n^{(3)}$ is the mean number of edges of a randomly chosen neighboring face of the typical face of a 3D tessellation under the condition that it has n edges, N is the random number of edges of the typical face, and $\mu_2 = Var(N)$. Note that in general $E(N) \neq 6$ but

$$E(N) = \frac{2\pi}{\pi - E(\rho)},$$

where ρ is the angle between two randomly chosen edges at the typical node. Unfortunately, $E(\rho)$ is not a universal constant for all 3D tessellations and neither is $E(N)$. The $E(N)$ in (24) for aluminum is 5.09 and μ_2 is 2.9 (Aboav, Private communication, 1994). It is interesting that in (24) there is no arbitrary constant a as in (5) for the 2D case.

Using Mecke's [1] Palm method, Chiu [27] has obtained an identity for 3D, which is similar to (18),

$$m_n^{(3)} = E(N) + \frac{E(N) - n}{2n} + \frac{cov[k(n,N),N]}{2np_n},$$

where $k(n,N)$ and p_n have the same meaning as before, except that they are defined for the typical face of a 3D tessellation. The 3D Weaire's sum rule is similar to that in 2D

$$\sum_n nm_n^{(3)}p_n = E(N^2).$$

LE CAËR'S TOPOLOGICAL LATTICE MODEL

Le Caër [59, 60] studied a topological *lattice* model that does not depend on geometrical considerations. Consider a lattice (say, a square lattice) in which every node, or vertex, is characterized by its coordination number of valence, z, which is the number of edges emanating from the node. Vertices of valence $z > z_s = d + 1$ in the d-dimensional space are unstable as their properties will be changed by small deformations (see also Rivier [4]). To remove the instability of a vertex of valence z, we add $z - d - 1$ edges to it. When $d = 2$, every added edge is connected to at least one of the other added edges if there are more than one edges added. This produces a set of possible stable configurations, called states, for the vertex of valence z; every state is characterized by a z-dimensional vector with components taking integer values within 1 and $z - 2$. Exact expressions for p_n and nm_n for this kind of lattice models have been obtained (see also Delannay, Le Caër, and Khatun [51]; Delannay, Le Caër, and Sfeir [61]; and Le Caër and Delannay [30]). This idea was based on Thompson [22]. An important application of this lattice model is in simulation studies (Rivier [4]). Moreover, experimental studies (described later) also showed that the topological properties of some natural froths are in good agreement with lattice models.

Experimental studies of cellular tissues in human amnion, epidermal epithelium of the cucumber, and Pb-Tl alloys, and simulations using 2D hard-disk models and models for liquid show satisfactory agreement in p_n with that calculated from the topological models associated with a distribution of independent states on a square lattice (Le Caër [59]). A deeper discussion for the epidermal epithelium of the cucumber and for the 2D hard-disk simulations can be found in Delannay, Le Caër, and Khatun [51]. Some more simulation results based on this topological lattice model using an Ising cellular structure were also reported in that study.

They also derived that if the $M_j(n)$ are linear in n, then the topological correlation functions, $A_{jn} \equiv M_j(n)/p_j$, are linear both in j and n owing to the symmetry of $A_{jn} = A_{nj}$ and are uniquely expressed as

$$A_{jn} = n + j - 6 - \frac{a}{\mu_2}(n - 6)(j - 6), \qquad (25)$$

where a is that parameter in (5); see also Delannay, Le Caër, and Sfeir [61]. They also suggested the use of these topological correlation functions A_{jn} for comparing topological properties of tissues with different distribution p_n.

Le Caër and Delannay [30] used grains of planar cuts of alumina polycrystals to calculate p_n, nm_n, and A_{jn}; and these values turned out to be similar to that obtained by exact calculations of the lattice model associated with a distribution of independent and equiprobable states on the vertices of tilings by triangles. But in Poisson Voronoi tessellations, only a similarity in A_{jn} between the simulated and the theoretic values was observed.

GODRÈCHE, KOSTOV, AND YEKUTIELI'S TOPOLOGICAL MODEL

Godrèche, Kostov, and Yekutieli [42] studied the following topological model.

Start from any ordinary equilibrium configuration of $C + 2$ cells. An edge is chosen randomly to perform the neighbor switching (T_1) transformation. Each cell configuration generated in this way corresponds to a planar Feynman diagram of a ϕ^3 field theory with a fixed number of vertices. Taking the limit $C \to \infty$, they derived analytically that

$$nm_n = 7n + 3 + \frac{9}{n + 1},$$

with $\mu_2 = 10.5$. This result was confirmed by their simulations. Approximately when n is large, we obtain a linear Aboav's law of the form (5) with $a \approx -1$.

However, any application of this model for modelling natural cellular networks was not suggested. One reason (Le Caër, private communication, 1994) may be that the 10.5 is much larger than any μ_2 observed in natural structures.

LEWIS' LAW—RIVIER'S MAXIMUM ENTROPY METHOD

Lewis [7, 8, 25, 62, 63] observed in several 2D cellular mosaics, at various stages of their

development, a specific relationship between the average area of a typical n-cell, A_n say, and n,

$$A_n = \alpha(n - 2), \tag{26}$$

where $\alpha = 1/(4\lambda_2)$ and λ_2 is the intensity of the point process of cell centroids.

Rivier and Lissowski [56] showed that *if there exists a linear relationship between A_n and n, for $n \geqslant 2$, then Lewis' law can be obtained.* Their argument was, however, not so rigorous because matrices of infinite rank were involved. Rivier [24, 4] gave a simple proof, namely, let $A_n = k_1 + k_2 n$ be true, where k_1 and k_2 are some constants. Because $E(N) = 6$ and $E(A_N) = 1/\lambda_2$ (expectation with respect to N), $k_1 + 6k_2 = 1/\lambda_2$. By writing $k_2 = \beta/\lambda_2$, where β is some constant, we obtain

$$A_n = \frac{\beta}{\lambda_2}\left[n - \left(6 - \frac{1}{\beta}\right)\right]. \tag{27}$$

Because a convex cell must have at least 3 edges, it is $A_2 = 0$, which yields $\beta = 1/4$. Hence, Lewis' law (26) is obtained.

Rivier and Lissowski, however, added the strange remark that the condition that a cell has at least three edges is too strong in general and stated that Lewis [25] and Smoljaninov [20] had examples of tissues following (27) with $2 \neq 6 - (1/\beta) < 3$. This may correspond to cases when nonconvex cells are allowed (see Introduction), or the linear relationship is not valid when $n = 2$. Indeed, there is no reason to assume that this linear relationship has an intercept at $n = 2$ (Rivier, private communication, 1993). Rivier [4] argued that β is actually the time that measures the ageing of a soap froth structure and a polycrystal, but this interpretation is not applicable to biological tissues.

Rivier and Lissowski then used the maximum entropy method to show that the linear relationship between A_n and n maximizes the entropy S under constraints (28), (29) and (30) below.

$$\Sigma p_n = 1, \tag{28}$$

$$\Sigma A_n p_n = \frac{1}{\lambda_2}, \tag{29}$$

$$\Sigma(6 - n)p_n = 0. \tag{30}$$

There may exist some hidden constraints.

Mombach, de Almeida, and Iglesias [52] discussed the maximum possible area of an n-edged cell with given perimeter and the absence of zero area cells. Some other topological constraints were already mentioned in the section about Rivier's microreversible system and maximum entropy method.

Rivier's later work [48] showed that if further physical constraint "the average energy of a cell is proportional to its perimeter" exists, which has been observed in metallurgical grain structures, the entropy S is maximized by the perimeter law or Desch's law,

$$\Pi_n = \alpha'(n - n_0'),$$

where α' and n_0' are some constants and Π_n is the mean perimeter of a typical n-cell. It then replaces Lewis' law.

It was found (e.g., Rivier [4]) that

$$\frac{\Pi_n}{A_n} = \frac{\pi}{L_n},$$

where L_n is the average chord length of a typical n-cell. Thus, Lewis' law and Desch's law can coexist in the special case that L_n is a constant. Some comparisons between Lewis' law and Desch's law in various froths can be found in Le Caër and Delannay [9].

However, the above studies were based on the maximum entropy method. Similar to Aboav's law, Chiu [50] showed that this maximum entropy method indeed cannot be used to prove Lewis' law nor Desch's law (see the section about Rivier's microreversible system and maximum entropy method and Appendix B).

SOME EMPIRICAL STUDIES OF LEWIS' LAW

Le Caër and Ho [45] and Drouffe and Itzykson [47] showed empirically that, for Poisson Voronoi tessellations, A_n varied linearly with n for $n < 11$. A change of the slope was observed by the latter authors for $n > 12$.

Lewis' law can be reexpressed as

$$A_n = \frac{1}{\lambda_2}\left(\frac{n - n_0}{6 - n_0}\right) \tag{31}$$

for some n_0. Crain [64] has suggested empirically that, for Poisson Voronoi cells, this n_0 is close to zero. (In his example this n_0 is about 0.4.) This result is, however, inconsistent with other studies. Other simulation values of n_0 for Poisson Voronoi tessellations were 1.4 (using the data in Quine and Watson [65]), 1.6 (using Le Caër and Ho [45]) and 1.7 (using Drouffe and Itzykson [47]). (Boots [66] has suggested that perhaps the columns for the conditional average second moment of the perimeter and for the area of a typical n-cell have been transposed erroneously in Crain's article. If this is true, then that n_0 in his example would be 1.4 instead of 0.4). For Voronoi cells generated by eigenvalues of a particular random matrix, Le Caër and Ho [45] has obtained that $n_0 = -1.78$, whereas for dynamic Voronoi cells immersed in a heat bath, Lauritsen, Moukarzel, and Herrmann's [49] simulations obtained $n_0 = 5.77$ for high temperature and $n_0 = 5.5$ for low temperature.

Quine and Watson [65] estimated the first four moments for the area and the perimeter of a typical Poisson Voronoi n-cell and conjectured that

$$A_n = \frac{2n - 3}{9\lambda_2}, \tag{32}$$

which is a special case of (27) or (31) with $\beta = 2/9$ or $n_0 = 3/2$.

The slope of (31) is $1/\lambda_2(6 - n_0)$ and $1/(6 - n_0)$ was calculated by Okabe, Boots, and Sugihara [3] (using least squares estimation) to be 0.200 (using Quine and Watson's [65] data), 0.219 (using Crain [64]), 0.229 (using Le Caër and Ho [45]) and 0.256 (using Drouffe and Itzykson [47]) for Poisson Voronoi tessellations. However, I recalculated the slopes, and the values are different from their calculation: 0.1993 (using Quine and Watson [65]), 0.2189 (using Crain [64], assuming that the columns for the conditional average second moment of the perimeter and for the area of a typical n-cell have been transposed; otherwise it is 0.1785), 0.2277 (using Le Caër and Ho [45]) and 0.2571 (using Drouffe and Itzykson [47]). Because there is an obvious change of the slope in Drouffe and Itzykson [47] for $n > 12$, and a linear relation fits A_n and n pretty well for

$n < 11$, I also calculated the slope for $n < 11$ and obtained 0.2270.

Brakke [68] used numerical integration to obtain the following second order moment: $Var(A) = 0.2802$, $Var(\Pi) = 0.9455$, $Var(N) = 1.7808$, $cov(A,\Pi) = 0.4905$, $cov(A,N) = 0.4009$ and $cov(\Pi,N) = 0.6506$, where A is the random area of a typical cell, and Π is its random perimeter. Thus, we can obtain the coefficients of correlation: $r(A,\Pi) = 0.9529$, $r(A,N) = 0.5675$ and $r(\Pi,N) = 0.5014$. Hinde and Miles [67] had already obtained almost identical results by simulation. Crain [64] found that in general $r(A,P|N = n)$ increases with n. Moreover, it was found empirically that the distribution of cell areas of planar sections of polycrystals and various Voronoi tessellations are fitted well by a gamma distribution (Le Caër and Delannay [9]).

Rivier [48] suggested a 3D version of Lewis' law (namely, the expected volume of a typical cell): under the condition that it has f faces, V_f,

$$V_f = b\left(\frac{\mathcal{V}_0}{\mathcal{F}_0}\right)[f - E(F)] + \frac{\mathcal{V}_0}{\mathcal{F}_0}, \tag{33}$$

whereas 2D Lewis' law can be reexpressed in a similar form,

$$A_n = b'\left(\frac{\mathcal{R}_0}{\mathcal{N}_0}\right)[n - E(N)] + \frac{\mathcal{R}_0}{\mathcal{N}_0}, \tag{34}$$

where F is the random number of faces of the typical cell of a froth, $\mathcal{V}_0$ ($\mathcal{R}_0$) is the total volume (area) of the froth, $\mathcal{F}_0$ ($\mathcal{N}_0$) is the total number of cells, and b (b') is a constant.

SOME SPECIAL MODELS STUDIED RECENTLY

Gervois et al. [38] and Lemaître et al. [39, 40] found that

$$A_n = an + b + \frac{c}{n},$$

where a, b, and c are some constants in the experiments of hard disks on an air cushion table. Moreover, it was found that A_n is not a monotonically increasing function of n, but attains its minimum at $n = 5$ or 6.

Mombach et al. [69] studied cellular networks with mitosis; a main difference between biological cells and cells of other froths is the existence of mitosis among biological cells. They found that Lewis' (as well as a linear Aboav's) law has an agreement with both simulation and experimental data. They also found empirically that the distribution of the cell area attains its maximum at non-zero area if mitosis is allowed, whereas if mitosis is forbidden, the maximum of the distribution is attained at zero area. They suggested that the existence of zero area cells should be suppressed by adding some more constraints. A theoretical treatment of a statistically equilibrated tessellation in which cell divisions are allowed can be found in Rivier, Arcenegui-Siemens, and Schliecker [70]. Delannay and Le Caër [35] simulated a 2D cellular structure in which cell divisions are allowed. The results of nm_n and A_{jn} (the topological correlation function) showed significant but small deviations from the linearity in n.

Flyvbjerg [71] suggested a nonlinear dynamic model that contains no free parameter, and all rates are determined dynamically. Using this model, he was able to derive the following form of Lewis' law (for this model only),

$$A_n = \alpha(n - 6) + \beta + O\left(\frac{1}{n}\right),$$

where α and β are known. Therefore, the linear Lewis' law is only asymptomatically valid in this model.

THE FUNDAMENTAL REGION

Zuyev [72, 73] introduced the notion of the fundamental region for a Voronoi polygon in the d-dimensional space.

Consider a cell generating point x and its corresponding Voronoi cell $T(x)$. For any vertex of $T(x)$, there are $d + 1$ equidistant nearest cell generating points, x, x_1, x_2, . . ., x_d. That is to say, there exists a ball centered at this vertex having x, x_1, . . ., x_d on its boundary and no other generating points

inside it. x or each x_i forming $T(x)$ lies in the intersection of d such balls. The union of all these balls for x and all x_i's forming $T(x)$ is called the *fundamental region* for $T(x)$ and is denoted by $D(x)$.

Russo's [74] formula for Bernoulli processes is modified to the case of Poisson processes and using this formula, Zuyev showed that the volume of the fundamental region of a typical Voronoi polygon, given n hyperfaces, follows a gamma distribution (see also Miles [75]). Because for any x, the volume of $T(x)$ does not exceed the volume of $D(x)$ divided by 2^d, a rough upper bound on the volume distribution of a typical Voronoi polygon having n hyperfaces is obtained.

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APPENDIX A
SOME EXCERPTS OF CHIU [27]

Weaire's proof of the sum rule (2) is "heuristic" because it involves a counting process not possible to perform as we have to count infinitely many cells and divide infinity by infinity. A more rigorous treatment of this sum rule is still needed. Chiu [27] studied this Weaire's sum rule and Aboav's law in the d-dimensional space ($\mathbf{R}^d$). The following is the particular case $d = 2$.

The proof may be a bit too mathematical. The monograph by Stoyan, Kendall, and Mecke [5] can serve as a useful background for this Appendix. Nevertheless, the materials below are self-contained.

The exact identity (18), although not very physically illuminating, helps us to show the fallacy of the maximum entropy method (see Appendix B).

NOTATION

Consider stationary planar tessellations that have only convex cells and are in *ordinary equilibrium state*, meaning there are always three edges emanating from each node.

Let $\mathbf{T} = \{\theta\}$ be the class of all such tessellations, where θ denotes such a tessellation. Let $C_0(\Theta)$ be the cell of the random tessellation Θ containing o, the origin, when o does not lie on an edge of Θ, and be the empty set otherwise. Let $\alpha_k(\Theta)$ be the sets of nodes, of edge-midpoints, and of cell-centroids of Θ for $k = 0, 1$, and 2, respectively. Let

$$\beta(\Theta) \equiv \bigcup_{k=0}^{2} \alpha_k(\Theta).$$

The intensities of these point processes $\alpha_k(\Theta)$ and $\beta(\Theta)$ are λ_k and λ', respectively, where λ' is positive and finite. If P is the distribution of a stationary random tessellation Θ, the *Palm* distribution of $\beta(\Theta)$, P', is

$$\lambda' P'(A) = \int \sum_{x \in \beta(\theta) \cap [0,1]^2} I_A(\theta - x) P(d\theta),$$

where A is the subclass of tessellations in $\mathbf{T}$ such that all tessellations in A have certain properties that are of interest and $I_A(\cdot)$ is the indicator function of A, that is, $I_A(\theta) = 1$ if $\theta \in A$ and zero otherwise. Clearly, the Palm distribution P' is also a distribution of tessellations. Suppose Θ follows this Palm distribution P'. When $o \in \alpha_2(\Theta)$, $C_0(\Theta)$ is called the *typical* cell (see Mecke [1], and Stoyan et al. [5]). This *typical* cell has the same distribution as a randomly chosen cell in the tessellation Θ, where all cells are equally weighted.

Let $\mathbf{T}_k$ be $\{\theta \in \mathbf{T} : o \in \alpha_k(\theta)\}$ and for any measurable function $g_k : \mathbf{T}_k \to [0, \infty)$; $E_k g_k(\Theta)$ is the mean of $g_k(\Theta)$ with respect to P', under the condition that $o \in \alpha_k$, i.e.,

$$E_k g_k(\Theta) = \frac{\lambda'}{\lambda_k} \int_{\mathbf{T}_k} g_k(\theta) P'(d\theta),$$

for $k = 0, 1$, and 2.

Moreover, let, for integer n, $\mathbf{T}_2^{(n)}$ be the set $\{\theta \in \mathbf{T}_2 : C_0(\theta) \text{ has } n \text{ edges}\}$. If $C_0(\theta) = \emptyset$, then let $C'_0(\theta)$ be the empty set, otherwise let it be the union of the closures of $C_0(\theta)$

and its neighboring cells; we call this union the *complex formed by* $C_0(\theta)$ and it is *typical* if $C_0(\theta)$ is the typical cell. Define $e(x,\theta)$ to be the number of edges of the cell containing x, where $x \in \alpha_2(\theta)$.

For random $\Theta \in T_2$, denote the random number of edges of the typical cell of Θ by $N \equiv N(\Theta)$ and the random total number of edges of the neighboring cells of the typical cell of Θ by $M^\Sigma \equiv M^\Sigma(\Theta)$. Let $p_n \equiv P\{N = n\}$. μ_2 denotes $Var(N)$, and $k(j,n)$ is the mean number of cells in $C_0(\theta)$ having j edges when $C_0(\theta)$ has n edges, where n is an integer such that $p_n > 0$, and zero otherwise. Thus, $k(j,N)$ is a random variable. A well-known and important result is $E_2(N) = 6$ for all planar stationary tessellations in ordinary equilibrium state.

For the case of presentation, in the text the subscript k in $E_k(\cdot)$ for $k = 1, 2,$ or 3 is omitted.

MECKE'S [1] THEOREM 4.2

For any measurable function $h : R^2 \times T \rightarrow [0, \infty)$,

$$\int \sum_{x \in \beta(\theta)} h(x,\theta) P'(d\theta) =$$

$$\int \sum_{x \in \beta(\theta)} h(-x, \theta - x) P'(d\theta).$$

WEAIRE'S SUM RULE

$$E(M^\Sigma) = E(N^2) = 36 + \mu_2. \tag{35}$$

Proof. Consider the following integral,

$$\lambda' \int_{T_2} \sum_{x \in \beta(\theta)} I_{T_2^{(n)}}(\theta) I_{\alpha_2(\theta) \cap C_0(\theta)}(x) e(x,\theta) P'(d\theta)$$

$$= \lambda_2 E\{I_{\{\Theta \in T_2 : N(\Theta) = n\}}(\Theta)[M^\Sigma(\Theta) + N(\Theta)]\}$$

$$= \lambda_2 \sum_i E\{I_{\{\Theta \in T_2 : N(\Theta) = n\}}(\Theta)[M^\Sigma(\Theta) + N(\Theta)]$$

$$|N(\Theta) = i\} p_i$$

$$= \lambda_2 E[M^\Sigma + N | N = n] p_n. \tag{36}$$

By Mecke's [1] Theorem 4.2, this integral is equal to

$$\lambda' \int \sum_{x \in \beta(\theta)} I_{T_2^{(n)}}(\theta - x) I_{\alpha_2(\theta - x) \cap C_0(\theta - x)}(-x)$$

$$e(-x, \theta - x) P'(d\theta). \tag{37}$$

The product of the indicator functions does not vanish if and only if $\theta \in T_2$ and $x \in \alpha_2(\theta)$ $\cap C_0(\theta)$ such that the cell containing x has n edges. Therefore, (37) is equal to

$$\lambda_2 E[Nk(n,N)] . \tag{38}$$

Because $\Sigma_n k(n,N) = \#$ cells in $C_0(\theta)$, given that $C_0(\theta)$ has N edges $= N + 1$. Hence, summation over all possible values of n of both (36) and (38) yields

$$E(M^\Sigma) + E(N) = \sum_n E[Nk(n,N)]$$

$$= E(N^2) + E(N).$$

Theorem. *When* $p_n > 0$, *we have*

$$m_n \equiv \frac{E(M^\Sigma | N = n)}{n}$$

$$= 5 + \frac{6}{n} + \frac{cov[k(n,N), N]}{np_n}. \tag{39}$$

Proof. From (36) and (38) we have the following identity,

$$E(M^\Sigma + N | N = n) p_n = E[Nk(n,N)]$$

$$= E(N) E[k(n,N)]$$

$$+ cov[k(n,N),N] . \tag{40}$$

If we can show that $E[k(n,N)] = (n + 1) p_n$, then the result follows. Consider the following integral,

$$\lambda' \int \sum_{x \in \beta(\theta)} I_{T_2^{(n)}}(\theta) I_{\alpha_2(\theta) \cap C_0(\theta)}(x) P'(d\theta) =$$

$$\lambda_2 (n + 1) p_n. \tag{41}$$

By Mecke's [1] Theorem 4.2, this integral is equal to

$$\lambda' \int \sum_{x \in \beta(\theta)} I_{T_2^{(n)}}(\theta - x) I_{\alpha_2(\theta - x) \cap C_0(\theta - x)}(-x) P'(d\theta).$$

$$\tag{42}$$

The product of the indicator functions does not vanish if and only if $\theta \in T_2$, $x \in \alpha_2(\theta) \cap C_0(\theta)$ and the cell containing x has n edges. Therefore, (42) is equal to $\lambda_2 E[k(n,N)]$. The result follows.

THE COVARIANCE TERM

As can be seen in Table 1, $cov[k(n,N),N]$ is empirically always less than μ_2. Moreover, $cov[k(n,N),N]$ is usually but not always positive; see Chiu [27].

Table 1. Simulated Poisson Voronoi tessellation, $\mu_2 \approx 1.726$ (Le Caër and Ho [42])

n	p_n	m_n	$\dfrac{cov[k(n,N),N]}{np_n}$	$\dfrac{cov[k(n,N),N]}{p_n}$	$cov[k(n,N),N]$
3	0.0113	7.009	0.009	0.027	0.0003
4	0.1068	6.718	0.218	0.872	0.0931
5	0.2595	6.492	0.292	1.460	0.3789
6	0.2946	6.315	0.315	1.890	0.5568
7	0.1986	6.171	0.314	2.197	0.4363
8	0.0905	6.050	0.300	2.400	0.2172
9	0.0295	5.948	0.281	2.532	0.0747
10	0.0074	5.859	0.259	2.590	0.0192
11	0.0014	5.780	0.235	2.580	0.0037

APPENDIX B
SOME EXCERPTS OF CHIU [50]

Chiu [50] showed that (22), $M_j(n) = A_j + B_j n$, is only a *sufficient but not necessary condition* for S to be maximized under the following constraints considered in Peshkin, Strandburg, and Rivier [37].

$$\sum_n p_n = 1, \tag{43}$$

$$\sum_n np_n = 6, \tag{44}$$

$$\sum_j M_n(j)p_j = np_n, \tag{45}$$

$$p_j M_n(j) = p_n M_j(n). \tag{46}$$

PESHKIN, STRANDBURG, AND RIVIER'S [37] MAXIMUM ENTROPY METHOD

Let us consider their argument again. They argued that if $M_j(n)$ is written in the linear form (22), then the constraint (45) can be re-expressed as a "linear" combination of the constraints (43) and (44) and so the entropy S can be increased further. (However, it is indeed not a 'linear' combination. This will be explained later.)

By summing up all possible j of both sides of (46), we find that the constraint (45) is a consequence of the constraint (46), which is also made redundant by the linear form (22); this has not been established in their article.

To show this redundancy of (46), note that

substituting (22) and (19) into the exact identity (18) yields

$$nm_n = 5n + 6 + \frac{cov(A_n + \delta_{nN} + B_n N, N)}{p_n}$$

$$= 6n + \frac{B_n}{p_n}\mu_2, \tag{47}$$

as $cov(\delta_{nN}, N) = (n - 6)p_n$. Compare (47) with (23) we obtain

$$\frac{B_n}{p_n}\mu_2 = \left(\sum_l lA_l\right) + \left(\sum_l lB_l\right)n - 6n, \tag{48}$$

for reasons that will become clear, the running index is changed from j to l.

By (43), (44), and (45), (22) becomes

$$A_l + 6B_l = lp_l. \tag{49}$$

Multiplying both sides of (49) by l and then summing up all possible l yields

$$\sum_l lA_l + 6\sum_l lB_l = \mu_2 + 36, \tag{50}$$

Substituting (50) into (48) yields

$$\frac{B_n}{p_n}\mu_2 = \mu_2 + (n - 6)\left(\sum_l lB_l\right) - 6(n - 6). \tag{51}$$

Therefore, from (22), (49), and (51),

$$\frac{M_n(j)}{p_n} = \frac{A_n + B_n j}{p_n}$$

$$= \frac{np_n + B_n(j - 6)}{p_n} \qquad \text{by (49)}$$

$$= n + \frac{(j - 6)}{\mu_2}\left(\mu_2 + (n - 6)\left(\sum_l lB_l\right) - 6(n - 6)\right) \quad \text{by (51)}$$

$$= n + j - 6 + \frac{(j - 6)(n - 6)}{\mu_2}$$

$$\left(\sum_l lB_l - 6\mu_2\right)$$

$$= \frac{M_j(n)}{p_j}.$$

Thus, (46) is fulfilled. [Compare with (25), we find that $a = \sum_l lB_l - 6\mu_2$.]

Note that the linear form (22) does not make the constraint (45) to be a linear combination of the constraints (43) and (44). A linear combination of, say $f(\mathbf{p})$ and $g(\mathbf{p})$, is in the form $af(\mathbf{p}) + bg(\mathbf{p})$ where a and b are constants, which are independent of $\mathbf{p}$. Now from (49), A_l and B_l depend on p_l and so (45) is not a linear combination of (43) and (44). However, because A_l and B_l are arbitrary, (49) does not impose a new constraint on $\{p_n\}$. Therefore, the maximum entropy prediction of $\{p_n\}$ is simply the solution obtained by maximizing $S \equiv -\sum_n p_n \ln(p_n/q_n)$ subject to the constraints (43) and (44). Let the solution be $\{p_n^{ME}\}$ and the maximum entropy value S^{ME}. Note that because the prior $\{q_n\}$ is unknown, it is not possible to obtain the explicit form of $\{p_n^{ME}\}$ nor the value of S^{ME}.

OTHER POSSIBLE MAXIMUM ENTROPIC PREDICTIONS

Let us reconsider this argument, using $k(j,n)$. Substituting (19) into the constraints (45) and (46) yields

$$\sum_j k(n,j)p_j = (n + 1)p_n, \quad (52)$$

$$p_j k(n,j) = p_n k(j,n). \quad (53)$$

If Peshkin, Strandburg, and Rivier's [37] arguments were applied on $k(j,n)$, we would conclude that $k(j,n)$ is also linear in n, i.e.,

$$k(j,n) = A_j' + b_j' n, \quad (54)$$

where A_j' and B_j' are some constants depend only on j. Although a linear Aboav's law can still be the result,

$$nm_n = \sum_j jk(j,n) - n$$

$$= \left(\sum_j jA_j'\right) + \left(\sum_j jB_j' - 1\right)n, \quad (55)$$

it is impossible to have both $M_j(n)$ and $k(j,n)$ being linear in n because they are different by 1 at the point $j = n$ and are the same otherwise. However, the entropy S in this case is still S^{ME}, because with the linear form (54), it can be shown easily that

$$\frac{k(n,j)}{p_n} = n + l - 5 + \frac{(j - 6)(n - 6)}{\mu_2}$$

$$\left(\sum_l B_l' - \frac{6}{\mu_2}\right)$$

$$= \frac{k(j,n)}{p_j},$$

and so (53), as well as (45), (46), and (52), is also redundant. Therefore, whether it is $M_j(n)$ or $k(j,n)$ (but not both) that is linear in n, the entropy is still S^{ME}, and so it is not clear which functional form is "more probable."

It is also possible to establish another entropic prediction of the form of $M_j(n)$ with the same entropy value S^{ME}, by making the constraint (45) to be really a linear combination of (43) and (44). Define

$$f(j,n) = M_j(n) - \delta_{nj} j = k(j,n) - \delta_{nj}(j + 1).$$

Then both constraints (45) and (52) can be rewritten as

$$\sum_n f(j,n)p_n = 0. \quad (56)$$

Using the argument of this maximum entropy method, when the constraint (56) is a linear combination of the constraints (43) and (44), then the entropy S can be increased further, when $f(j,n)$ is in the form,

$$f(j,n) = a_j + b_j n,$$

for some constants a_j and b_j depending on j only, then (56) is a linear combination of (43) and (44), provided that $a_j + 6b_j = 0$ (which does not depend on $\{p_n\}$). Thus, $M_j(n)$ and $k(j,n)$ are nonlinear,

$$M_j(n) = (b_j + \delta_{nj})n - 6b_j,$$
$$k(j,n) = (b_j + \delta_{nj})n - 6b_j + \delta_{nj}.$$

Surprisingly, a nonlinear Aboav's law is obtained,

$$nm_n = \sum_j jM_j(n) = n^2 + \left(\sum_j jb_j\right)n - 6\sum_j jb_b,$$

and $M_j(6) = 6\delta_{6j}$, which means that all neighbors of the typical 6-cell are also cells of 6 edges! It means that p_6 is either zero or 1! Although it seems that such a froth is unlikely to be a real equilibrated natural structure, this structure has the same entropy value as those with the two linear Aboav's laws considered before, namely, S^{ME}. This can be proved by using a similar argument as above.

Actually, whenever $\sum_j M_j(n) = n$ and $M_j(n)/p_j$ is a function that is symmetric in n and j, (46) holds and so does (45). Thus, it may be helpful to use the topological correlation function $A_{jn} \equiv M_j(n)/p_j$, introduced by Delannay, Le Caër and Khatun [51], Delannay, Le Caër, and Sfeir [61], and Le Caër and Delannay [30]. Then, (45) and (46) can be rewritten as

$$\sum_j A_{nj}p_j = n,$$

$$A_{nj} = A_{jn}.$$

Now, suppose that A_{nj} is $(nj)/6$, $1 + (n - 1)(j - 1)/5$, $2 + (n - 2)(j - 2)/4, \ldots$, or $n\delta_{nj}/p_n$, $1 + (n - 1)\delta_{nj}/p_n, \ldots$ etc. (see also equation (19) of Le Caër and Delannay [30]. Then, the constraints (45) and (46) are redundant. Hence, the entropy can still be maximized to S^{ME}; that is to say, all these forms are "equally probable." That means $M_j(n)$ is not uniquely determined by this maximum entropy method, nor are A_{jn} and $k(j,n)$.

CONCLUSIONS

The fallacy of this method is due to that the maximum entropy is always S^{ME}, no matter

how the constraints (45) and (46) are made redundant. Thus, $M_j(n) = A_j + B_{jn}$ is only a *sufficient but not necessary condition* for $S = S^{ME}$.

It is not yet clear which $M_j(n)$, $k(j,n)$, $f(j,n)$, A_{jn}, and/or some other characteristic(s) their/its functional form(s) can be chosen arbitrarily. Even if we can set up some criteria such that only the functional form of $M_j(n)$, say, can be chosen arbitrarily, it is not necessary to be linear. Even if it had to be linear, the slope (B_j) and the intercept (A_j) would not be fixed. Thus, the structure can undergo changes without changing the value of the entropy and so the structure is not in statistical equilibrium.

Hence, the maximum entropy method can only be used to obtain the following:

a linear Aboav's law

$\Updownarrow$ S is maximized

$\Updownarrow$ statistical equilibrium

However, the example presented above shows that this method cannot be used to derive a unique linear Aboav's law (nor Lewis' law). The equivalence between the statistical equilibrium and the maximum value of S is in doubt because the structure can undergo changes without reducing or increasing S. Thus, the following is *wrong*:

a unique linear Aboav's law

$\Uparrow\!\!\!\!\!\!\!/$ S is maximized

$\Downarrow\!\!\!\!\!\!\!/$ statistical equilibrium

Indeed, the example above shows that

S is maximized $\Longrightarrow$ infinitely many possible forms of Aboav's law
$\Longrightarrow$ not necessary in statistical equilibrium.