Supplementary Information

1. Thermodynamic Entropy

1.1. Mass and Energy Budget for an Open System in Non-Thermal Equilibrium

For open large systems experiencing non-thermal equilibrium it is possible to define an elemental volume where we can assume local equilibrium. Both intensive and extensive variables can be defined at this local domain. On the one hand, the intensive quantities such as temperature and pressure can be defined direclty. On the other hand, it is posible to define the extensive variables by implementing the respective densities. Therefore, the concept of elemental volumes and local equilibrium allows us to define all thermodynamics variables as a function of space and time [1].

The mass balance for a component k in an elemental volume is given by:

$$\frac{dn_k}{dt} + \nabla \cdot \frac{\mathbf{I}_k}{M_k} - \sum_j \nu_{j,k} v_j = 0. \tag{S1}$$

In this equation the n_k refers to number of mols of component k per unit volume, $\mathbf{I}_k$ refers to the flux of component in [mass/area/time], M_k is the molar mass of component k in $[\frac{\text{Mass}}{mol}]$, $\nu_{j,k}$ is the stoichiometric coefficient for component k in the reaction j, and v_j is the velocity of the reaction j. The term $\sum_j \nu_{j,k} \xi_j$ is a net sink (or source) term of n_k over all possible reactions where k is involved within the elemental volume. Note, that total mass is conserved, and the sink (or source) term of a component k implies that there is a source (or sink) in other components.

The energy balance equation for an elemental volume is given by:

$$\frac{de}{dt} + \nabla \cdot \mathbf{J}_e = 0 \tag{S2}$$

where $\mathbf{J}_e$ is the total flux of energy in the elemental volume. We consider three main components for $\mathbf{J}_e$: (i) a flux of kinetic energy $\mathbf{J}_c$, (i) a flux of energy that is directly related with the internal energy of the control volume $\mathbf{J}_u$, and (ii) a flux of energy in the form of radiation.

$$\mathbf{J}_e = \mathbf{J}_c + \mathbf{J}_u + \mathbf{J}_{rad}. \tag{S3}$$

Under a gravitational field $\mathbf{J}_c$ and $\mathbf{J}_u$ can be expressed as:

$$\mathbf{J}_c = \sum_k \frac{1}{2} M_k \mathbf{v}_k^2 \mathbf{I}_k \tag{S4}$$

$$\mathbf{J}_u = \mathbf{J}_q + \sum_k \mathbf{I}_k \left(u_k + M_k g z \right) \tag{S5}$$

where M_k is mass of component k, u_k is the internal energy in component k, z is the gravity, and g is the gravity.

1.2. Entropy Budget for Open Systems in Non Thermal Equilibrium

The entropy budget of an elemental volume is given by:

$$\frac{ds}{dt} + \nabla \cdot \mathbf{L}_s = \sigma \tag{S6}$$

where $\mathbf{L}_s$ is the flux of entropy, and σ is the entropy production in the elemental volume. Note that entropy is not conserved. In this document we divide σ in two:

$$\sigma = \sigma_{int} + \sigma_{rad}. \tag{S7}$$

where σ_{int} refers to the production of entropy within the elemental volume associated with transformations of different forms of energy to heat, and σ_{rad} is the production of entropy associated with the dissipation of incoming fluxes of radiation in other forms of energy at the surface.

It can be shown that for an elemental volume experiencing a gravitational field, and receiving radiative fluxes of energy the flux of entropy $\mathbf{L}_s$ is given by:

$$\mathbf{L}_s = \frac{\mathbf{J}_q}{T} + \sum_k s_k \frac{\mathbf{I}_k}{M_k} + \mathbf{L}_{rad} \tag{S8}$$

where $\mathbf{L}_{rad}$ is the flux of entropy associated with radiation fluxes, and s_k is the molar entropy associated with component k which is defined as:

$$s_k = \frac{u_k - \mu_k}{T} \tag{S9}$$

where u_k is the molar internal energy in component k under the absence of a gravitational field, and μ_k is the molar chemical potential associated with component k. Similarly, it can also be shown that the entropy production σ_{int} is given by:

$$\sigma_{int} = \mathbf{J}_u \cdot \nabla \left(\frac{1}{T} \right) - \sum_k \frac{\mathbf{I}_k}{M_k} \cdot \nabla \left(\frac{\mu_k}{T} \right) + \sum_j \frac{A_j^f}{T} v_j$$

$$- \sum_k \frac{\mathbf{I}_k}{T} \cdot \nabla(gz) + \frac{\delta\phi}{T} \tag{S10}$$

In this equation the term A_j^f is the affinity of the reaction j, ϕ is the dissipation rate of kinetic energy to heat in the elemental volume, and $\mathbf{J}_u$ is the energy flux in the absence of a gravitational field.

$$\mathbf{J}_u = \mathbf{J}_q + \sum_k \frac{\mathbf{I}_k}{M_k} u_k \tag{S11}$$

Equations (S7) and (S10) describe the production of entropy for an elemental volume exposed to fluxes of radiation and experiencing a gravitational field. Note that there are five terms on the right side of Equation (S10). Each of these terms refer to a particular process, (i) *heat transfer*; (ii) *difussion*; (iii) *chemical reactions*; (iv) *loss of potential energy under a gravitational field*; and (iv) *dissipation of kinetic energy*.

1.3. Entropy Generation

If we integrate in space over the total volume of the open system and if we define some time scale τ we can compute the total entropy generated ($\dot{S}_{gen}$) in the system during these spatial and time scales as:

$$\dot{S}_{gen} = \int\limits_0^\tau \int\limits_V \frac{ds}{dt} dV dt + \int\limits_0^\tau \int\limits_V \nabla \cdot \mathbf{L} dV dt \geq 0 \tag{S12}$$

By applying the divergence theorem it is possible to express $\dot{S}_{gen}$ in terms of the fluxes across the control volume boundary.

$$\dot{S}_{gen} = \int\limits_0^\tau \left(\frac{dS_{T,cz}}{dt} \right) dt + \int\limits_0^\tau \int\limits_{A_{cz}} \mathbf{L} \cdot \hat{n} dA dt \geq 0 \tag{S13}$$

where $dS_{T,cz}$ is the total instantaneous change of entropy in the control volume, and $\hat{n}$ refers to the unit normal vector field to A_{cz}. Using Equation (S8) we can obtain the next expression for the $\dot{S}_{gen}$:

$$\dot{S}_{gen} = \int\limits_0^\tau dS_{T,cz} + \int\limits_0^\tau \int\limits_{A_{cz}} \left(\frac{\mathbf{J}_q}{T} + \sum_k s_k \frac{\mathbf{I}_k}{M_k} + \mathbf{L}_{rad} \right) \cdot \hat{n} dA dt \geq 0 \tag{S14}$$

1.4. Calculation of Entropy at Steady State

At steady state the rate of change of entropy at the elemental volume is negligible, thus from Equation (S6):

$$\nabla \cdot \mathbf{L} = \sigma. \tag{S15}$$

In some situations the instantaneous change of entropy at elemental volumes within the system is not negligible ($ds/dt \neq 0$), but over a period of time τ the total change of entropy in the control volume could be considered negligible:

$$\int\limits_0^\tau dS_{T,cz} \approx 0 \tag{S16}$$

This particular scenario is useful in the Critical Zone (CZ) where the annual change of entropy could be considered low enough to be to be neglected. In this case the generation of entropy can be computed based on the fluxes of entropy across the surface only:

$$\dot{S}_{gen} = \int\limits_0^\tau \int\limits_{A_{cz}} \mathbf{L} \cdot \hat{n} dA dt \geq 0 \tag{S17}$$

2. Derivation of $\sigma_{cz,int}$

This section shows the derivation for the internal production of entropy σ in the presence of the Earth's Gravitational Field for an elemental volume that experiences fluxes of mass and heat with the environment only, radiation is neglected. This derivation is based on Kondepudi and Prigogine [1] Chapter 15.

The rate of change in energy e for an elemental volume where local equilibrium is assumed can be stated as:

$$\frac{\partial e}{\partial t} = \frac{\partial u}{\partial t} + \frac{\partial c}{\partial t} \tag{S18}$$

where u is the internal energy and c is the kinetic energy. The rate of change of energy is also given by:

$$\frac{\partial e}{\partial t} = -\nabla \cdot \mathbf{J}_e \tag{S19}$$

where $\mathbf{J}_e$ is the total flux of energy into the control volume:

$$\mathbf{J}_e = \mathbf{J}_q + \sum_k (u_k + c_k + M_k g z)\mathbf{I}_{Mk} \tag{S20}$$

where $\mathbf{J}_q$ is the heat flux, and u_k is the mean molar internal energy content in component k. Term $c_k = 1/2 M_k \mathrm{v}_k^2$ is the mean molar kinetic energy in component k where $\mathrm{v_k}$ is the mean velocity of component k. The balance of kinetic energy can be expressed as:

$$\frac{\partial c}{\partial t} = -\nabla \cdot \underbrace{\sum_k (c_k \mathbf{I}_{Mk})}_{\mathbf{J}_c} - \delta\phi \tag{S21}$$

where $\mathbf{I}_{Mk} = \mathbf{I}_k / M_k$ is the molar flux of component k, and $\delta\phi$ is a sink term in this equation that reflects the dissipation of kinetic energy due to transformation to heat. We can use the previous expressions to compute the rate of change in u in the elemental volume as:

$$\frac{\partial u}{\partial t} = \frac{\partial e}{\partial t} - \frac{\partial c}{\partial t} = -\nabla \cdot \underbrace{\left(\mathbf{J}_q + \sum_k (u_k + M_k g z)\mathbf{I}_{Mk}\right)}_{\mathbf{J}_u^o} + \delta\phi \tag{S22}$$

where J_u^o is the flux related to u under the Earth's Gravitational Field.

The Gibbs relation is given as:

$$T ds = du - \sum_k \mu_k dn_k - \sum_k M_k g z\, dn_k. \tag{S23}$$

Based on Equation (S23) the rate of change of entropy can be expressed as:

$$\frac{\partial s}{\partial t} = \frac{1}{T}\left(-\nabla \cdot \mathbf{J}_u^o + \delta\phi\right) + \sum_k \left(\frac{\mu_k + M_k g z}{T}\right)\left(\frac{\partial n_k}{\partial t}\right) \tag{S24}$$

Note that:

$$\frac{\partial n_k}{\partial t} = -\nabla \cdot \mathbf{I}_{Mk} - \sum_j \nu_{j,k} v_j \tag{S25}$$

where v_j is the velocity of a reaction j and $\nu_{j,k}$ are the stoichiometric coefficients for component k in a reaction j. It can be shown from Equation (S24) that:

$$\frac{\partial s}{\partial t} + \nabla \cdot \left[\frac{\mathbf{J}_u^o}{T} - \sum_k \left(\frac{\mu_k + M_k g z}{T}\right)\mathbf{I}_{Mk}\right] = \mathbf{J}_u^o \cdot \nabla\left(\frac{1}{T}\right) - \sum_k \mathbf{I}_{Mk} \cdot \nabla\left[\frac{\mu_k + M_k g z}{T}\right] + \sum_j \frac{A_j^f v_j}{T} + \delta\phi \tag{S26}$$

where A_j^f is the affinity of the chemical reaction j. We can define a flux $\mathbf{J}_u$ that does not include the fluxes related to a gravitational field:

$$\mathbf{J}_u^o = \underbrace{\mathbf{J}_q + \sum_k u_k \mathbf{I}_{Mk}}_{\mathbf{J}_u} + \sum_k M_k \mathbf{I}_{Mk} \tag{S27}$$

also note that:

$$\sum_k \mathbf{I}_{Mk} \cdot \nabla\left(\frac{M_k g z}{T}\right) = \sum_k \frac{\mathbf{I}_{Mk} M_k g}{T} \nabla z + \sum_k I_{Mk} M_k g z \cdot \nabla\left(\frac{1}{T}\right) \tag{S28}$$

Using Equations (S27) and (S28) we can express Equation (S26) as:

$$\frac{\partial s}{\partial t} + \nabla \cdot \left[\frac{\mathbf{J}_q}{T} - \sum_k \left(\frac{s_k}{T}\right)\mathbf{I}_{Mk}\right] =$$
$$\left(\mathbf{J}_q + \sum_k u_k \frac{\mathbf{I}_k}{M_k}\right) \cdot \nabla\left(\frac{1}{T}\right) - \sum_k \frac{\mathbf{I}_k}{M_k} \cdot \nabla\left(\frac{\mu_k}{T}\right) + \sum_j \frac{A_j^f v_j}{T} + \sum_k \frac{\mathbf{I}_k}{T} g \cdot \nabla(z) + \frac{\delta\phi}{T} \tag{S29}$$

where s_k is the molar entropy content of component k. Comparing Equation (S26) with the entropy budget of the local control volume experiencing a flux of entropy $\mathbf{L}$ associated with heat and mass fluxes only:

$$\frac{\partial s}{\partial t} + \nabla \cdot \mathbf{L} = \sigma_{prod} \tag{S30}$$

We observe that the term to the right in Equation (S29) represents the entropy production within the local domain.

$$\sigma_{prod} = \underbrace{\left(\mathbf{J}_q + \sum_k u_k \frac{\mathbf{I}_k}{M_k}\right) \cdot \nabla\frac{1}{T}}_{\text{Heat Flux}} - \underbrace{\sum_k \frac{\mathbf{I}_k}{M_k} \cdot \nabla\frac{\mu_k}{T}}_{\text{Diffusion}} + \underbrace{\sum_i \frac{A_j^f v_j}{T}}_{\text{Chemical Reactions}}$$
$$- \underbrace{\sum_k \frac{\mathbf{I}_k}{T} g \cdot \nabla(z)}_{\text{Potential Energy Transformation}} + \underbrace{\frac{\delta\phi}{T}}_{\text{Kinetic Energy Transformation}}. \tag{S31}$$

3. Thermodynamic Entropy of Radiation

Radiation is an main form of heat transfer. A proper understanding and computation of the entropy associated with radiation is needed to have a complete quantification of the entropy budget in the CZ. In this document we show the most important equations. For more details see Wu and Liu [10].

With the introduction of the quantum hypothesis Max Planck was able to obtain an expression for the energy (J_ν), and entropy (L_ν) fluxes of a blackbody radiation beam at a particular frequency ν:

$$J_\nu = \frac{h\nu^3}{c^2}\left(\frac{1}{\exp(\frac{h\nu}{\kappa T} - 1)}\right), \tag{S32}$$

$$L_\nu = \frac{h\nu^2}{c^2}\left\{\left(1 + \frac{c^2 J_\nu}{h\nu^3}\right)\ln\left(1 + \frac{c^2 J_\nu}{h\nu^3}\right) - \left(\frac{c^2 J_\nu}{h\nu^3}\right)\ln\left(1 + \frac{c^2 J_\nu}{h\nu^3}\right)\right\}. \tag{S33}$$

In these equations, c is the speed of light, h is the Planck constant, and κ is the Boltzmann constant. The radiation entropy $\mathbf{L}_{rad}$ flux emitted from a blackbody over a surface can be obtained through integration over the solid angle, and frequencies:

$$\mathbf{L}_{rad} = \int\limits_{o}^{\infty} \int\limits_{\Omega} L_{\nu} cos\theta d\Omega d\nu, \tag{S34}$$

where θ is the zenith angle. It can be shown that;

$$\mathbf{J}_{rad} = \sigma_b T^4_{source}, \tag{S35}$$

and

$$\mathbf{L}_{rad} = RF \, \sigma_b T^3_{source} = RF \frac{\mathbf{J}_{rad}}{T_{source}}. \tag{S36}$$

where T_{source} is the temperature of the body emitting the radiation, σ_b is the Stefan-Boltzmann constant which has been obtained both experimentally and theoretically. It can be shown that [10]:

$$\sigma_b = \frac{2\pi^5 \kappa^4}{15 c^2 h^3}. \tag{S37}$$

Term RF in Equation (S36) is the radiation factor (usually $RF > 1$). Note that the RF differentiates the formulation for the computation of entropy in radiation and heat fluxes. The value of RF for blackbody radiation is $4/3$. However non-blackbody radiation is ubiquitous on Earth. In order to quantify the energy and entropy fluxes of non-blackbody radiation Petela [11] introduced the concept of a perfect gray body that behaves as a blackbody but the spectral radiation energy flux is equal to the Planck function for blackbody radiation affected by an emissivity ε. Assuming that ε is frequency independent it is possible to show that the energy flux of non-blackbody radiation is $\mathbf{J}_{rad} = \varepsilon \sigma T^4$. However, the same does not hold for the entropy flux. In other words, even under the assumption that ε is frequency independent the entropy flux of non-blackbody radiation $\mathbf{L}_{rad} \neq \frac{4}{3} \varepsilon \sigma_b T^3$. In order to compute the entropy flux of a non-blackbody radiation we must integrate $L_v(\varepsilon)$ over the solid angles and frequencies ν:.

$$\mathbf{L}_{rad} = \int\limits_{o}^{\infty} \int\limits_{\Omega} L_{\nu}(\varepsilon) cos\theta d\Omega d\nu. \tag{S38}$$

However, this integration is challenging as $L_{\nu}(\varepsilon)$ is non-linear. Landsberg and Tonge [12] developed an approximate solution of Equation (S38) by considering a diluted blackbody radiation with a dilution factor δ, which can be considered analogous to the emissivity ε. This approximation has shown good results except for low values of ε. A more recent approximation developed by Wright *et al.* [13] using a similar approach as Landsberg and Tonge [12] was able to overcome this problem, and has shown good accuracy in the solution of Equation (S38). Both Landsberg and Tonge [12] and Wright *et al.* [13] formulations have been very helpful to quantify the fluxes of entropy of radiation in processes related to the CZ.

4. Description of Optimum Principles

4.1. Minimum Work (or Energy) Rate Expenditure

An initial optimum hypothesis based on minimum work was suggested by Murray [22,23] for physiological systems. In this case *work* was related with a cost of operation that could be associated with system maintenance. In the CZ a first approach related with work was performed by Leopold and Langbein [24] that proposed the principle of least work for streams. According to Leopold and Langbein [24] this principle is compatible with the principle of minimum entropy production. Although the study by Leopold and Langbein [24] was criticized for its lack of formalism in the application of thermodynamic concepts, it established the foundation of a new paradigm to analyze the geomorphological evolution of the CZ. Different principles based on minimum work, energy, or power were postulated to understand the landscape evolution. These principles were able to capture the patterns observed in meanders, the formation or riffles and pools, and the formation of self-similar river networks [25]. There has been a long debate about the validity of these principles and their application in fluvial geomorphology. Today, the debate still continues and the validation with experimental information and models is not definitive.

The connection between these minimum principles with other principles associated with thermodynamic entropy that have been applied in the CZ is still unclear. An interesting analogy was performed by Molnar and Ramirez [26] with the implementation of the Gyarmati principle to analyze the flow in open channels. The Gyamarti principle proposed the maximization of a function G_y defined as:

$$G_y = \left(\int\limits_V (\sigma - \psi)dV \right) \tag{S39}$$

where σ is the local entropy production and ψ is the local dissipation potential rate. Note that function G_y involves a space integration of the entropy production and therefore this principle is not equivalent with the maximum entropy production principle (MEPP) because MEPP is a local based principle. Molnar and Ramirez [26] were able to show and equivalence between the Gyarmati principle and the minimum energy dissipation rate principle under some special conditions of river flow.

4.2. Minimum Entropy Production Principle, MinENT

The minimum entropy production principle MinENT was postulated by Prigogine [27]. According to Prigogine [27] systems in the linear non-equilibrium regime reach a steady state ($\frac{dS}{dt} = 0$) at which the entropy production from the system ($\int\limits_V \sigma dV$) is constant and attains a minimum value [1]. Note that the objective function in this principle refers to the total production of entropy that involves a space integration of the local entropy production σ. This principle can be applied only for the linear regime and provides information only at the stationary state. In recent years this principle has been criticized mainly because it does not provide sufficient further insight about the system beyond to what is already known from balances of mass, momentum, and energy [28,29].

4.3. Maximum Power Output Principle, MPOP

The maximum power output principle MPOP was initially proposed by Odum and Pinkerton [30]. They suggested that systems tend to maximize the power output. As explained in Odum and Pinkerton [30], the power output in this principle refers to the definition of power implemented in thermodynamics which is the rate of energy transfer by work, or useful energy. According to this principle, systems tend to use useful available energy, become more efficient, and therefore maximize the power output. According to this hypothesis in order to maximize power there has to be a trade off between the efficiency (useful work produced) and rate. Processes with high efficiency occurs very slow, while fast processes end up producing less useful work and destroying free energy in the form of heat. The maximun power output would occur at an intermediate point between these two conditions. Hall [31] introduces interesting examples in biochemistry, economy, car engines, and coal burning where this principle occurs. From a human perspective this principle would be a condition that any ideal system would pursue. For instance, the goal of human made systems is usually to maximize work per unit of time. Although this principle is very logical, the main question is whether nature is designed with this objective? Additionally, the computation of actual power for a system such as the CZ is challenging and could become arbitrary at some point. Power output is a quantity that is difficult to quantify as it goes beyond the second law of thermodynamics [32]. Concepts such as exergy provides a limit about the potential work that can be extracted instead of the actual work that is performed by the system. Some approaches have used a similar formulation as exergy to calculate the limits of maximum work that can be extracted from a flux of exergy for different processes of the CZ [33,34].

The MPOP was reformulated by Odum [35] in terms of maximizing emergy. According with Odum [35] more power output could be stated in terms of a maximization of emergy. The principle was restated again by Odum [36] in terms of maximization of empower, and was proposed by Odum as the fourth law of energetics. There is no decisive evidence of the maximum power principle [37]. However, its supporters argue it is challenging to test this principle and it could take some time similarly as it has taken with previous ecological hypotheses including Darwin's natural selection [31]. However, there is some indirect evidence that nature choose intermediate states in efficiency and rate, at which power attains a maximum. One example of intermediate states is the leaf area index of deciduous forest which is maintained at an intermediate state where it produces a maximum power.

4.4. Maximum Entropy Production Principle MEPP

Open systems in a state that is far away from equilibrium experience a continuous production of entropy sustained by a constant incoming flux of energy with low thermodynamic entropy from the environment. This production of entropy allows these systems to maintain the structure and organization, and increase the amount of information. Therefore, it is likely that if there is an optimum function pursued by open systems as they move away from equilibrium, it will be associated with thermodynamic entropy. However, the question is whether these systems maximize thermodynamic entropy? On the one hand some authors suggest that open systems optimize rather than maximize entropy production [38]. On the other hand some authors support the maximum entropy principle (MEPP) that suggest there is a maximization at a local scale in these systems as the move away from equilibrium.

The MEPP was initially proposed by Ziegler [39,40]. According to this principle when thermodynamic forces (F_i^T) are preset, the fluxes are such that maximize the entropy production density at the local scale $\sigma(J_i^T)$ [28]. This principle seems a promising fundamental alternative that determines the behavior of open systems in far from equilibratebrium conditions. There are some experimental examples that support this hypothesis (See Manuscript). The optimization problem to find J_j^T can be stated as:

$$\text{maximize:} \qquad \sigma(J_i^T)$$

$$\text{subject to:} \quad \sigma(J_i^T) = \sum_i F_i^T J_i^T. \qquad\qquad \text{(S40)}$$

and can be solved with the next lagrangian function [29]:

$$La = \sigma(J_k) - \lambda\left(\sigma(J_k^T) - \sum_i F_i^T J_i^T\right) \qquad\qquad \text{(S41)}$$

where λ is a Lagrange multiplier. The thermodynamic fluxes J_k^T that maximize σ can therefore be determined by $\frac{\partial La}{\partial J_j^T} = 0$. As Martyushev [29] pointed the relationship between thermodynamic forces and fluxes may be both linear and nonlinear. This feature makes this principle very powerful. In addition, the principle holds for non-stationary state conditions, and it is possible to derivate MinENT from Equations (S40) and (S41) by implementing appropriate conditions [29].

Note that the objective function and the principle itself is conceptualized at a local domain. The entropy production from the total system by integration of the local entropy production may not be maximum. According to Martyushev [29] previous studies have erroneously refuted the principle by performing calculations of entropy production over the entire system through spatial integrations. However, several studies have also provided support for the principle from computations of total entropy over the entire system. Therefore, implementations linking this principle with the total entropy production from a system that is computed from a space integration of the local entropy production is not appropriate. In particular, it is important to keep in mind the local formulation of MEPP in the CZ where most of the applications are performed over large control volumes. However, the local formulation can be very useful to infer general properties of the system at local scales and then use these properties to analyze the system at larger scales.

Production of entropy involving larger spatial and time scales are more linked with the Entropy Generation Maximum Principle (EGMa) proposed by Lucia [41]. This principle deals with the generation of entropy and represent an interesting alternative that is more feasible to be tested.

4.5. Principle of Maximum Exergy Storage, PMES

This principle was originally proposed by Jørgensen and Mejer [42–44] and has been analyzed in latter publications [45,46]. A continuous flux of exergy is responsible for driving a system towards a state that is far from thermodynamic equilibrium. Therefore, it is very likely that exergy is an important variable that controls the evolution of open systems in a state that is far from equilibrium. According to this hypothesis the most probable path that a system exposed to exergy fluxes will take as it moves far and far from equilibirum is that in which it maximizes the storage of exergy. In the case of ecological systems

eco-exergy is used instead of exergy. This principle has been tested on several ecological systems [47–49] and seems a good goal that guide the development of ecosystems. Analysis of exergy storage captures the 24 properties of ecosystem maturity proposed by Odum [50] as indicators of maturity [51]. Although the goal of this principle is related to a "*storage*" of exergy, this may result in similar patterns as those obtained if maximum power is maximized, since maximization of exergy storage could enhance the generation of power [52]. However, exergy only poses a maximum limit on the availability of useful energy. Therefore, maximization of exergy storage will provide only an alternative where the system becomes effective in maximizing the storage of potential useful energy that can be used to perform work. It does not assure that the system is maximizing the work that is produced because exergy could be destroyed in different ways including some where no work is produced. Therefore, PMES and MPOP are different hypotheses and it needs to be proven that both leads to the same patterns in all conditions.

4.6. Principle of Maximum Exergy Dissipation, PMED

This principle was originally proposed by Schneider and Kay [53,54] and implies that as open thermodynamic systems are moved away from equilibrium due to external fluxes of exergy, these systems will respond by organizing themselves and degrading exergy as much as possible. In addition, as the systems are moved further away from equilibrium they will enhance more the organization patterns and become more effective to degrade exergy. Degradation of exergy is connected with generation of entropy. Therefore, this principle is similar with a maximum generation of entropy (EGMa) Lucia [41]. Similarly to PMES a main motivation in the conceptualization of this principle is the connection of exergy with useful energy and work. However, as mentioned above exergy only poses a limit on the availability of useful energy to perform work. Therefore, dissipation of exergy not necessarily means maximization of work per unit time.

4.7. Principle of Maximum Energy Flow

In his seminal paper *Contribution to the Energetics of Evolution*, Alfred Lotka suggested that natural selection and therefore evolution tends to maximize the energy flux through the system in accordance with the constraints that are imposed. This paper represented an important contribution to the energetics of ecology and even today, the implications of this study are still under consideration. There have been some attempts to connect Lotka's principle with others that are more recent. However, as explained below this connection could be analyzed from different angles that may be adjusted accordingly with different principles.

Lotka states that the units of the objective that he proposed to maximize are *power*. Therefore, it has been suggested that this principle is connected with the MPOP. However, it can be arguable what exactly Lotka meant when he used the word *energy*. It seemed that energy as used in his paper refers to a form of useful energy to perform work and therefore could be associated with exergy. Also, according to Lotka if other things are equal the energy flux will be proportional to the mass in the system. This mass is referred in his paper as organic matter and therefore his objective of energy flux will result similar to maximizing eco-exergy storage as proposed in PMES [37]. However, there are some constraints on mass production based on available resources. According to Lotka under such constraints there will be a maximum power

from the turnover of available organic matter. In this case the turnover of organic matter (eco-exergy) could be associated with exergy dissipation by the system as suggested by the PMED.

4.8. Principle of Maximum Entropy POME

This principle was initially proposed by Jaynes [55,56]. According to this principle when making inferences from data that is incomplete the probability distribution that should be selected is the one that satisfies the maximum information entropy subject to given constraints. This principle is motivated on the fact that distributions with higher entropy are more probable. The optimization problem can be stated as [14]:

$$\text{maximize:} \qquad H(X)$$

$$\text{subject to:} \qquad \int\limits_a^b f(x)dx = 1 \qquad\qquad (S42)$$
$$\int\limits_a^b g_r(x)f(x)dx = \overline{g_r(x)}$$

where the first constraint results from the definition of a probability densify function, and the other constraints are given by particular properties of the system under consideration and are defined in terms of moments. According with Singh and Rajagopal [14,57] this maximization problem can be solved with Lagrange multipliers with the next lagrangian function

$$La = -\int\limits_a^b f(x)\text{Ln}(f(x))dx - (\lambda_0 - 1)\left(\int\limits_a^b f(x)dx - C_0\right)$$
$$- \sum_{r=1}^n \lambda_r\left(\int\limits_a^b f(x)g_r(x)dx - C_r\right). \qquad (S43)$$

In this equation, λ_r refers to the lagrange multiplier that is used for constraint r. The function $f(x)$ that satisfies the maximum $H(X)$ can then be obtained from $\frac{\partial La}{\partial f} = 0$. This principle has been widely used in hydrology to infer the probabilistic distribution of different variables. However, it has not been used as a predictive principle that guides the evolution of open systems that are far from equilibrium. Dewar [58,59] tried to link this principle with MEPP but his approach has been criticized by Bruers [29,60].

4.9. Principle of Maximum Ascendency

Ascendency is defined for networks as the product of total system throughout and average mutual information ($As = IT$). Ascendency has been correlated with the 24 properties proposed by [50] as indicators of maturity. Based on this evidence Ulanowicz [61,62] proposed a principle that will help to understand ecosystem development. According to this principle, in the absence of major perturbations, ecosystems will exhibit a tendency to increase in ascendency [63]. This principle is particularly important because it is directly associated with the network organization of the ecosystem which is a major property that in some cases is underestimated. In addition, this is the only optimum principle from

all those described in this study where a concept directly linked with information entropy is included to conceptualize the development of ecosystems.

5. Multi-Objective Optimization

A multi-objective optimization problem can be stated as [64]:

$$\text{maximize:} \quad \varsigma(\mathbf{x})$$

$$\text{subject to:} \quad g_i(\mathbf{x}) \leq 0$$
$$h_i(\mathbf{x}) \leq 0$$

(S44)

where $\varsigma(\mathbf{x}) = (\varsigma_1(\mathbf{x}), \varsigma_2(\mathbf{x}), \varsigma_3(\mathbf{x}), ..., \varsigma_n(\mathbf{x}),)$ is a vector of optimum functions defined over a n-dimensional decision variable vector $\mathbf{x} = (x_1, x_2, x_3, ..., x_n)$, and $g_i(\mathbf{x})$, $h_j(\mathbf{x})$, $i = \{1, .., m\}$ $j = \{1, .., p\}$, represents all the constraints that must be fulfilled. The solution of this optimization problem is a Pareto front, that represents a trade off between all the objective functions that are considered. It represents the optimum points that are attained for an objective in such a way that it can not be optimized more without degrading some other objective values.

List of Symbols

Symbol	Description
SW	Shortwave
LW	Longwave
LE	Latent heat
HH	Sensible eat
I	Flux of mass
J	Flux of energy
L	Flux of entropy
$\mathbf{L}_{rad}$	Flux of entropy in the form of radiation
S	Thermodynamic entropy
W	Work as defined in thermodynamics
G	Free Energy
H	Information (Shannon) entropy
T	Temperature
g	Gravitational field
$\mathbf{V}_{cz}$	Control Volume to delineate the Critical Zone
A_{cz}	Surface area of the control volume in V_{cz}
$M_{T,cz}$	Total Mass in the CZ defined by V_{cz}
$E_{T,cz}$	Total Energy in the CZ defined by V_{cz}
$S_{T,cz}$	Total Entropy in the CZ defined by V_{cz}
$E_{ex,cz}$	Total Exergy in the CZ defined by V_{cz}

Symbol	Description
m_{cz}	Local mass in Critical Zone
e_{cz}	Energy of an elemental volume within the Critical Zone
s_{cz}	Entropy of an elemental volume within Critical Zone
M_k	Molar mass in component k
σ_{cz}	Entropy production by the control volume V_{cz}
$\sigma_{cz,rad}$	Local entropy production associated with a transformation of radiation
$\sigma_{cz,int}$	Local entropy production associated with the production of entropy from internal processes within the V_{cz}.
$\dot{S}_{gen,cz}$	Entropy generated by the CZ
$\dot{S}_{gen,cz,rad}$	Entropy generated by the CZ associated with instantaneous transformation of radiation at the surface
$\dot{S}_{gen,cz,int}$	Entropy generated by the CZ associated with internal processes within the CZ.
$\Gamma_{gen,cz}$	Destruction of exergy by the CZ
$\Gamma_{gen,cz,rad}$	Destruction of exergy by the CZ associated with instantaneous transformation of radiation at the surface
$\Gamma_{gen,cz,int}$	Destruction of exergy by the CZ associated with with internal processes within the CZ.
τ	Time scale associated with a particular process used to compute $\dot{S}_{gen,cz}$.
μ_k	Chemical potential in component k
n_k	Mole number per unit volume of component k
s_k	Molar entropy in component k
u_k	Internal energy in component k
c_k	Average kinetic energy associated with component k
h_k	Enthalpy associated with component k
A_i^f	Affinity of a chemical reaction
v_i	Velocity of a chemical reaction
$\nu_{j,k}$	Stoichiometric coefficients of component k in reaction j
$\delta\phi$	Amount of kinetic energy transformed to heat
RF	Radiation factor for computation of thermodynamic entropy of radiation
I_{PPT}	Mass flux associated with precipitation
I_{ET}	Mass flux associated with evapotrasnpiration
I_{ELV}	Mass flux associated with physical denudation
I_{GEO}	Mass flux associated with chemical denudation
J_{SW}	Energy flux associated with shortwave radiation
J_{LW}	Energy flux associated with longwave radiation
$J_{Q,LE}$	Energy flux associated with latent heat
$J_{Q,H}$	Energy flux associated with sensible heat
J_{ELV}	Energy flux associated with physical denudation
J_{GEO}	Energy flux associated with chemical denudation
$J_{Q,G}$	Ground heat flux

Symbol	Description
$J_{Q,atm}$	Flux associated with the heat absorbed by the atmospheric part of the critical zone.
J_{bio}	Energy flux associated with living systems in CZ.
J_{pho}	Energy flux associated with SW taken during photosynthesis.
$J_{Q,resp}$	Energy flux within the CZ associated with heat fluxes from respiration and dissipation of work of biological systems.
J_{Q,H_2O}	Energy flux within the CZ associated with heat fluxes from dissipation of kinetic and potential energy of water.
J_{H_2O}	Net energy flux associated with water fluxes.
L_{SW}	Entropy flux associated with SW radiation
L_{LW}	Entropy flux associated with LW radiation
$L_{Q,LE}$	Entropy flux associated with LE heat
$L_{Q,H}$	Entropy flux associated with HH heat
L_{ELV}	Entropy flux associated with physical denudation
L_{GEO}	Entropy flux associated with chemical denudation
$L_{Q,G}$	Ground heat flux
$L_{Q,atm}$	Entropy flux associated with heat absorbed by the atmospheric part of the critical zone.
L_{bio}	Entropy flux associated with living systems in CZ.
L_{pho}	Entropy flux associated with SW taken during photosynthesis.
$L_{Q,resp}$	Entropy flux associated with heat fluxes from respiration and dissipation of work of biological systems.
L_{Q,H_2O}	Entropy flux associated with heat fluxes from dissipation of kinetic and potential energy of water.
L_{H_2O}	Net entropy flux associated with water fluxes.
R_n	Net radiation
L_G	Entropy flux related to the ground heat flux
L_{SW}	Entropy flux related to shortwave radiation flux
s_{solar}	Solar constant of first order
e_{solar}	Solar constant of second order
σ_{evap}	Solar evaporation
$\dot{S}_{gen,atm}$	Entropy generated by a control volume in the atmosphere defined by V_{atm}
ρ_{water}	Water density
RHa	Relative humidity of air at temperature T_a
RHs	Relative humidity of air at saturartion at T_a
R_v	Gas constant for water vapor
g	Earth's Gravity
R	Gas constant
F_s	Variable that includes the free energy of the entire soil after scaling up from minerals.

Symbol	Description
S_s	Variable that includes the thermodynamic entropy of the entire soil after scaling up from minerals.
ΔF	Change in the free energy of the entire soil.
ΔS_s	Change in the thermodynamic entropy of the entire soil.
$E_{ex,CMInet}$	Net Exergy flux through the Critical Zone. It does not include exergy destruction
$\mu_{i,o}$	Chemical potential of a given component (or mineral) at a given reference.
Ecx	Ecoexergy.
$Escx$	Specific Ecoexergy.
c_i	Concentration of component i.
$\dot{Q}_{perc}$	Rate of heat produced through percolation
$\dot{Q}_{surf}$	Rate of heat produced through surface water flow
T_s	Soil temperature
T_{surf}	Temperature surface water
T_{source}	Temperature of body emitting radiation
T_o	Temperature of hypothetical environment that surrounds the CZ
σ_{soil_per}	Entropy production associated with the percolation of water in soils.
ζ_{str}	Discharge
$f(x)$	Probabilistic density function of random variable X
$p(x_i)$	Probability of occurrence of event x_i
$p(x_i, y_j)$	Probability of occurrence of event x_i and y_j
$H(X)$	Information (Shannon) Entropy of a random variable X
$H(X,Y)$	Join information entropy of random variables X and Y
$I(X,Y)$	Mutual Information of variables X and Y
$D_{KL(p\|\|q)}$	Kullback $-$ Leibler divergence between distribution p and q
Φ_n	Non symmetric overhead in ecological networks
Φ_s	Symmetric overhead in ecological networks
CMI	Conditional Mutual Information
DIT	Directional information transfer
TE	Transfer Entropy
F	Total system throughput
$F(i,j)$	Quantum flux leaving compartment i and entering compartment j
$F(i)$	Quantum flux leaving compartment i
$F(j)$	Quantum flux entering compartment j
B	Total system mass
B_i	Mass in system compartment i
B_j	Mass in system compartment j
D	Dependency
Rel	Relevance
Red	Redundancy

Symbol	Description
Ω	Number of possible arrangements of the microstates
J^T	Thermodynamic flux
F^T	Thermodynamic force
λ	Lagrange multipliers
La	Lagrangian functions
σ_b	Stefan-Boltzmann Constant

Conflicts of Interest

The authors declare no conflicts of interest.

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