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# Near-resonant diurnal reactions: a physical model applicable to origin of life processes

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## Abstract

We adopt that the large residual chemical potential energy (CPE) among reagents of the biosphere constitutes the key physical problem posed by life. We associate the formation of this CPE with the near-resonant behaviour of a two-stage ‘A-B’ molecular process that behaves as a self-sustaining parametric oscillator. Under suitable conditions, such an oscillator generates CPE when forced by a periodic (daily) insolation. The net growth factor required to explain the current mean excess of biospheric CPE is  $\sim 5 \times 10^{-12} \text{ d}^{-1}$ . This aligns with the mean exponential coefficient of secular oxygen generation in the terrestrial atmosphere. It is also consistent with a feasible scale of oxygen production in certain prebiotic natural photosynthesis scenarios, that can be candidates for the initial A subprocess on the Earth. We schematize initial evolutionary development of the A-B process, including the important role of the intermediate AB compound that provides negative feedback. Supportive C-type molecules also develop as a by-product. The diurnally related distribution of  $\text{H}_2\text{O}_2$  on Mars may illustrate a comparable proto-biospheric scheme, and there may be analogous processes on Jupiter. The exponential growth in the lengths of terrestrial nucleotide chain molecules also supports its validity, as does the corresponding growth in measures of cellular complexity. We compare the scenario’s implications with biological evidence on the possible co-evolution of blue-light photoreception and circadian timing in Archean photoautotrophs. We consider how a surviving level of cellular organization of circadian rhythmicity, from ancient through to modern times, may be interpreted along these lines, comparing our model with a previously published, comparable, biochemical one.

**Key Words:** Origin of life, chemical potential energy, parametric oscillator equation, insolation, biosphere, resonance, feedback, oxygen generation, protonucleosides

## 1. Introduction

The original impetus for this article was the problem of what defines life [1, 2, 3]. Further ramifications of the definition of life have been presented by Gleiser [4] as a far-reaching challenge with possible cosmological implications. Our response is collected in Section 2, which, in Section 3, leads into considerations of the energization of the terrestrial biosphere. We view this issue as central, and it strongly influences our approach. Section 4 recalls physical principles for a mechanism whereby potential energy can be steadily gained by a system. We consider possible manifestation of this in very early, biologically accepted, life forms. Section 5 sketches a generalized scenario for the realization of diurnal parametric oscillation at a proto-biotic level, before looking at cyanobacteria of the genus *Oscillatoria*. Finally, Section 6 makes a few practical points for future empirical testing. A deeply entrenched biotic rhythmicity continuing from the early terrestrial environment through to the present may become verifiable observationally.

The multi-disciplinary and fundamental nature of the subject makes for difficulties of presentation. Remarks that look elementary to specialists from one discipline may still be unfamiliar to those of another. However, the Darwinian character of evolution points to the origin of life reducing to something simple, despite the extreme complexity of present-day recognized forms. This point encourages hope that a broad-based reductionism can make progress with this deep problem.

## 2. The energization of living matter

The inertial principle embodied in Newton's first law of motion is a cornerstone concept of physics and relates the concept 'no action at a distance'. Material bodies do not suddenly display motion without some connection to an energy source. Given such kinetic energy, a body tends to degrade—via non-conservative interactions with its surroundings or within itself—into random motions of constituent particles or electromagnetic emission, dissipating its coherent kinetic energy into heat. This then radiates away so that a body, after an initial limited energy input, later comes to relative rest (equilibrates) with its surroundings.

Kinetic energy associated with the bulk motion of bodies in nature can usually be associated with a corresponding potential energy. In a mechanically conservative regime these two forms may continue to transform one into the other during motion, but in the presence of dissipative processes the total mechanical energy diminishes. Dissipation of the ordered motion implies insufficient subsequent energy to return the body to its former potential (for details, cf., e.g., [5]). States of (relative) rest are thus associated with potential energy minima.

There is something basic in distinguishing among lifelike matter. Even at deepest rest, for example, living matter remains potentially active. Some degree of organized capability to act—in short, via the mechanism of *metabolism*—is observed in all cases described as *alive* ([6, 7, 8, 9]). Since such coherent activity continues in time, we should consider its integrated propulsion, that is the directed energy of living matter, keeping in mind the foregoing conservation principle. The continuous energization of living matter in the face of heat losses in a freely dissipative environment thus implies a continuous supply of power directed specifically to maintain the condition of living [10].

From a materialistic viewpoint,<sup>1</sup> we can look to one basic source of external power associated with the life of the terrestrial biosphere, namely the energy radiated by the Sun. The solar flux amounts to around 1.36

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<sup>1</sup>This takes a minimalist approach. It is recognized that such a view may be unsatisfactory, or not be able to address all aspects of life, but such aspects lie outside the scope of this article.

kW for each normally projected square metre above the Earth's atmosphere. The total incident power over the whole Earth is then around  $2 \times 10^{14}$  kW or  $\sim 7 \times 10^{21}$  J per day. There may be other driving powers, related to the Earth itself or perhaps some other external sources, but noting the salient facts, such as the location of living matter about the Earth and the central role of photosynthesis within biological activity [11], it is natural to look primarily to the solar flux as the main physical driver. However, it is far from clear just how, in detail, the ordered energization of living matter is related to this influx.

Energy consumption associated with the basic metabolism of a human being, for example, is of order  $\sim 10^7$  J per day. For the totality of living humans, this implies a daily consumption of about  $6 \times 10^{16}$  J simply to remain alive. Of course, whilst a particularly energetic and numerous form, humans are just one of dozens of millions of living species. A reasonable estimate for the total power driving the entire biosphere then works out at  $\sim 10^{12}$  kW (cf. e.g. [12]). The solar flux is thus able to drive this 'engine', the problem becomes just how it works in practice. To say that the living processes of the biosphere are simply a redistribution of part of the solar flux is not enough; there are other processes driven in this way, for example the atmospheric weather, or marine currents. Such processes are energized mechanically and have enough inertia to maintain their motions for some short time in the absence of direct irradiance, but we would not normally characterize them as living. With the latter, something more than a dissipative mechanical system can be seen, although just where, in essence, the difference lies and how it has arisen, are not obvious.

### 3. Biospheric chemical potential energy

We can visualize the interplay of potential and kinetic energies at the microscopic level via the arrangements of charged particles within atoms or molecules. In particular, the existence of chemical bonds implies a chemical potential energy (CPE), since a bond requires there be a restoring force, or potential derivative, conserving relevant atomic structures in their bonded conditions. At the microscopic level, chemical potential, in the sense we concentrate on herein, is essentially the electrostatic potential associated with the relative excitations of valence electrons.<sup>2</sup> Chemical reactions are mostly accounted for by the changing configurations, or energy states, of such electrons. If, as a result of some anisotropy, exothermic reactions occur, particulate potential energy tends to diffuse irreversibly into electromagnetic radiation. An endothermic process implies an atypical initial particle configuration for a given environmental temperature, which becomes regularized into a more probable arrangement. In either case, physical results are comparable to the degradation of macroscopic motions. Under normal conditions, naturally occurring processes tend toward configurations whose typical valence electron energies tend toward those at the ambient temperature of the local environment (i.e. a Boltzmann distribution, [13]).

But this is not the case in the surface regions of Earth. There is a large residual CPE among the reagents of the biosphere, whose existence is the main problem we address. This CPE includes that of all currently living species, as well as formerly living material now resident in the upper layers of the Earth, i.e. all carboniferous oils, shales and coal measures. This energy can be (conservatively) estimated from ecological data, taking into account combustive yield, to be of the order of  $10^{22}$  J. This begins to compare with the total thermal energy of the Earth's atmosphere of  $\sim 10^{24}$  J, or  $\sim 1.5 \times 10^5$  J·kg<sup>-1</sup>, while the average energy density of biotic matter is now some  $3 \times 10^7$  J·kg<sup>-1</sup> [14, 15, 16].<sup>3</sup> The ratio of these two energy densities,  $\sim 200$ , is something we return

<sup>2</sup>Physically comparable potentials may also occur with other, generally weaker, inter-particle forces associated with, for example, molecular polarity, or crystalline structures.

<sup>3</sup>Measured energies associated with chemical processes usually relate to Gibbs free energy. This allows for some proportion of

to in the next section. But we can also approach the same issue by noting the existence of  $\sim 3.714 \times 10^{19}$  moles of free oxygen in the Earth's atmosphere, and comparing this with a more feasible initial planetary atmosphere made up of lower chemical potential constituents such as carbon dioxide, water, methane etc. Methane-oxygen combustion yields some 572.2 J/mole [17], so that a reverse process of oxygen liberation can be interpreted as a net potential gain of  $\sim 2 \times 10^{23}$  J. The very significant comparability of this datum to the potential energy yield of all organic matter around the Earth's surface has already been pointed out by Haldane [18] and the point was noted by Urey [19] in his account of the biosphere's energization.

This disequilibrium is not what one would expect according to normal thermodynamics. Typical chemical reactions occurring naturally near standard temperature and pressure conditions about the Earth's surface (e.g. firedamp ignition) would involve such short interaction times that excess molecular energies would be swiftly radiated away or lost through kinetic collisions with other molecules, until approximate equilibrium with the normal thermal energy of the surroundings is achieved. Life on earth is thus generally recognized to be quite distant from a regular condition of thermal equilibrium [13]. We presume, in this paper, this excess CPE has come originally from the solar radiated input that has been converted by living forms. Some of the CPE excess can remain inside a formerly living organism even when no longer functioning as a singly organized system in the way we consider below. This seems evident, for example, when living organisms consume other biotic material for food. But this, by itself, does not explain how the biospheric CPE excess continues to be accumulated.

It should be noted here that presently recognized life-forms on the Earth are in cellular arrangements. If we particularize the foregoing generalities to the compositions of the cellular fluids of organisms and the mechanisms regulating them, we see that in no instance is the chemical composition of a biological solution the same as that of its surrounding medium. Most organisms are in ongoing processes of gaining or losing water, and all cells continuously maintain steady solute concentration gradients between themselves and their external medium. This state of disequilibrium between cellular solutions and their surroundings requires significant energy expenditure.

The disparity between the compositions of biological solutions and surrounding environments shows up another point: cells do not accumulate solutes in direct proportion to their availability. These differences in cellular composition are reflected in the evolutionary progression of species, with varied solute concentrations giving rise to correspondingly evolved microenvironments. If we list the many different types of compounds present in the small volume of a cell, the issue of solute differentiation comes to the fore. How does the cell package so many different metabolites, enzymes, osmolytes, nucleic acids, and so on, into such a small space? Alternatively, what, other than highly non-uniform structures of localized and concentrated molecular energy, can account for cellular make-up?

## 4. Restorative behaviour and resonance

A living system shows a characteristic tendency towards 'homeostasis'; a restorative behaviour 'attracted to' continued living (e.g. [1, 20]). Most explanations of biological behaviour refer either directly or indirectly to this property; through its role in cell metabolism or implications for evolution. Let us now focus on the restorative property underlying homeostasis.

Homeostatic restoration can be associated with evolution theory, which interprets the features of given life

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energy invested in (entropic) changes of state that do not directly define the local photon field distribution (temperature). The total CPE prior to such a process would be greater than, but generally of the same order as, the Gibbs free energy, on general equipartition grounds.

forms as being necessary, more or less, for their continuation. Particular features within species are discussed in terms of how they affect chances of survival. Contemporary expositions refer to molecular arrangements within genes or gene combinations that determine such somal behaviour. Moreover, the future survival of species involves adaptation to progressive environmental changes. That certain species continue to survive verifies the ‘fitness’ of their development, i.e. the robustness of their special homeostasis.

This point permits teleological explanations of living behaviour. Absence of the apparent purpose of survival in a general environment would relax behaviour towards non-survival: its presence is therefore supportive, necessary even; but it has not been shown that only seeking survival is *sufficient* to ensure it. We argue that the evolving, restorative character of living behaviour in a dissipative environment involves an external energization.

Perhaps the most concise way to describe restorative behaviour is with a second order differential equation that can be written in a general way as  $\Delta^2\{y\} = 0$ , where the second-order operator  $\Delta^2$  acts on the variable  $y$ , dependent on some other variables  $x_i$ , and involves its rate of change  $D\{y\}$ , and the rate of change of  $D\{y\}$ , i.e.  $D^2\{y\}$ .<sup>4</sup>

More generally, if

$$\Delta^2 y = f(t), \tag{1}$$

where  $f(t)$  is some external agency causing a disturbance with a frequency component  $\Omega$ , it can be shown that the oscillation’s responsive amplitude  $y_0$  at the driving frequency  $\Omega$  satisfies

$$y_0^2(\Omega) \propto f_0^2 / \{(\Omega - \omega)^2 + (\lambda\Omega)^2\}, \tag{2}$$

where the driving function’s amplitude is  $f_0$ . Equation (1) now includes the dissipative term  $\lambda D\{y\}$ , that decreases the oscillatory behaviour in dependence on the damping coefficient  $\lambda$ . This quantity determines the ‘bandwidth’ of the response function  $y_0(\Omega)$ . The sharpness of the frequency response, or ‘ $Q$ -factor’, is inversely related to  $\lambda$ . Heavily damped oscillators have broad bandwidths, and respond to a wider range of driving frequencies around the resonance centre  $\Omega = \omega$ .

A more generalized form of restorative equation has coefficients that are not constant but have a (small) variation with time. This form can be written as

$$D^2\{y\} + \lambda(t)D\{y\} + \omega^2(t)y = 0. \tag{3}$$

This is sometimes known as the parametric oscillator equation. Solution to this equation can be shown to yield an amplitude that depends on  $t$  in the form

$$y_0(t) = y_{01}e^{(\alpha-\mu)t}, \tag{4}$$

in which  $y_0(t)$  grows or declines according to growth rate  $\alpha$  referenced to mean dissipation term  $\mu$ . The value of  $\alpha$  near the central resonance relates to the coefficient  $f_0$  of the driving term, now taken over to the left side and appropriately scaled into the term  $\omega^2(t)$ .

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<sup>4</sup>This is often written as  $d^2y/dt^2$  (and similarly with  $D \equiv dy/dt$ ). A comprehensive introduction to the physics of restorative systems can be found in, for example, [21] (Ch. 14). Parametric resonance is discussed in [22] (pp 462–471) and a useful guide to practical methods of dealing with relevant differential equations is that of [23] (especially part B2). The very simple form, involving the single independent time variable  $t$ ,  $D^2\{y(t)\} + \omega^2y(t) = 0$ , implies that the force acting at the displacement  $y$ , proportional to the accelerative term  $D^2\{y\}$ , must be restorative, since it increases in proportion to  $y$  but in the opposite direction. It is well-known that a form of  $y$  satisfying the foregoing linear equation is the oscillatory sinusoid  $y = a \sin(\omega t + b)$ , where  $a$  and  $b$  are constants and  $\omega$  is the frequency of the oscillation.

Specifically, for settled, near-resonantly driven conditions,

$$\alpha = f_0 \omega^2 \{1 - (\Omega^2 - (2\omega)^2)/f_0^2 \omega^4\}^{1/2}/4\Omega. \quad (5)$$

Note that the responsive cycle's timescale should be about half that of the driver for an efficient parametric oscillator ( $\Omega \approx 2\omega$ ). The dissipation term can be shown to be [23]

$$\mu = (\Omega/4\pi) \int_{2\pi/\Omega} \lambda(t) dt. \quad (6)$$

The dissipation depends on the available behaviour of the system, i.e. in what ways it can lose potential energy. In the present context, this concerns biospheric activity subject to a periodic (daily) influx of solar radiation. If we suppose that the CPE of a biochemically responsive system, that can be regarded as a parametric oscillator subject to a daily energy input  $f(t)$  from the Sun, scales with the amplitude  $y_0(t)$  as in equation (4), then the current mean CPE excess of  $\sim 200$  (Section 3, second paragraph) implies that  $10^{12}(\alpha - \mu) \approx \ln 200$ , or

$$(\alpha - \mu) \approx 5 \times 10^{-12} \text{ (d}^{-1}\text{)}. \quad (7)$$

The exponential character of this general growth should be noted, even its rate is very small.

Consider now the growth of oxygen in the Earth's atmosphere [24, 25], from the Earth's formation, through the Archean and Proterozoic periods to the present.<sup>5</sup> We find the average slope of the logarithmic growth to be  $\sim 4.30\text{--}4.8$  By, which when translated into the exponential growth per day amounts to  $\sim 5 \times 10^{-12}$ , which is remarkably close to the foregoing estimate in (7), keeping in mind the approximate nature of the calculation. We should also note that this agreement is reflected in the comparability of the CPE of the biosphere regarded as its presently combustible total energy and the energy required to liberate the mass of free oxygen in the Earth's present atmosphere from likely initial constituents noted in the preceding section. If we associate liberation of bonded oxygen with CPE enhancement then there is direct support for the resonating oscillator analogy to biosphere formation and development. We shall produce a scheme for this in the following section where we also argue for an appropriate estimate of the driving amplitude coefficient  $f_0$ .

We may note that the system of equations used to analyse biospheric variables (biomass, nutrient content, internal energy density etc.) discussed by, for example, Kondratyev et al. [26] are of essentially the same form as our highly condensed equation 3; and indeed damped oscillatory behaviour is demonstrated in the results of such analyses, although ecological interests generally concern longer periods than circadian.

The geophysical characteristics of the Earth thus set the 'boundary conditions' associated with the foregoing formulation's application to the physiology of living organisms. Changes in these conditions over geological timescales can be associated with changes characterizing life's emergence and development. Notable, for our present context, is the Earth's present sidereal day of about  $23^{\text{h}} 56^{\text{m}}$  is much slower than the possible  $\sim 6^{\text{h}}$  rotation period, when biotic processes may have been first starting [27, 28], though the year, by contrast, has remained more or less constant [28]. Fossil evidence may be used to check these points. In this way, very early life forms have been shown to involve molecular components associated with clock-like genetic functions, cycling proteins, and photoreceptors [29]. The comparison of such sequences through genetic analysis of biological clocks offers an important insight into life's early forms and evolution. That a circadian rhythmicity pervades

<sup>5</sup>An interesting summary diagram on this was shown by L. Gardiner of University Corp. Atmospheric Research at <http://www.windows.ucar.edu/> (2004).



the biosphere is not in doubt, but we propose a fundamental significance to the whole biospheric energization related to it. This can be understood by interpreting circadian rhythmicity, physically, as a quasi-resonant mechanism driven, at a basic level, by the diurnal insolation.

The Earth's early atmosphere is generally reckoned to have originally been a moderately reducing one, wherein the present level of atmospheric oxygen was bound largely as water, carbon dioxide or other mineral oxides [19, 30]. Urey [19], in fact, argued that the high CPE of such a postulated atmosphere, relative to what is currently found, could account for the CPE excess of life. That is an argument comparable to that of this paper in energy terms, except that we discuss how such a transfer could be maintained as a single developing physical process in the face of continuous dissipation and heat loss. Experiments in the 1950s showed that under conditions thought to simulate primitive Earth amino acids and simple organic acids could be produced [31, 32], although it is also now generally accepted that some assumptions of the early experiments were inappropriate, particularly the implied timescale for the oxidative transformation [33]. Nevertheless, the polymerization of large molecules from smaller subunits by dehydration and phosphorylation still appears basic for the required components of living forms. The first primitive RNA-like molecules may have somehow arisen from prebiotic precursors ('PNA' [34]). Note that the Moon was closer to the Earth during its early history and the Earth's rotation much faster. Earth should then have experienced more rapid and drastic cycles of solute dilution and re-concentration, with likely salt-dependent association/dissociation conditions. Under such conditions, multiplication of stable nucleic acid-like polymers could have occurred in a manner analogous to the cycling growth of polymerase considered by Lathe [27].

## 5. Molecular configuration (Schematic)

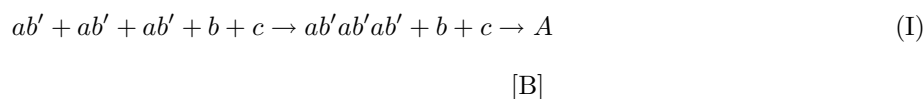
In what follows, we schematize a solar-driven molecular process that can correspond to a parametric oscillator, according to the foregoing. To allow for generality, apart from a few specific points mentioned later, chemical reactions are not detailed, though underlying physical principles can still be specified. We follow the classical Carnot concept of a two-stage cycle, including A and B subprocesses, in a way that may have some points of comparison with the thermosynthetic model of Muller [35, 36]. A Carnot cycle of some kind appears requisite to the generation of mechanical energy, as shown particularly in animals. But analogous processes are implied whenever energy is transformed from a photon source into forms available for biologically useful work. More general ideas about the role of cycling in biology and its relationship to currently known metabolic pathways are given, for example, by Morowitz et al. [37]. The biochemical model for a circadian bacterial oscillator of Pavlidis & Kauzmann [38] is also noteworthy in this context, and we later offer a closer comparison with that. Our presentation is more elementary, however, having the intention of addressing a prebiotic molecular ensemble, from which a continually developing biosphere evolves (for further general background cf. e.g. [30, 39, 40]).

Although, in reality, numerous alternative pathways may occur in the relevant chemical reactions, we schematize only the biophysical essentials. The issue of branching side-reactions was discussed by King [41] with a formulation comparable to the one we follow below. King gave a necessary condition on the ratio of side-reactions to promoting reactions that should be satisfied in the milieu in which an autocatalytic sequence can be maintained. He did not, however, demonstrate that this condition was sufficient. In any case, while the scheme we outline appears primarily directed towards the sugar-base combination of a simple nucleoside, it could, in principle, apply at higher levels. Harvey [42], for example, notes that isolated RNA is an essentially unstable compound, achieving stability only by protein binding. The underlying issue is one of suitably occasioned

molecular binding energy decreases (potential energy increases) that make available work used to support or enhance stability of the autocatalysis.

Let us suppose a terrestrial molecular envelope containing inorganic molecules of type a, b, c, d, etc. These molecules can be considered as fairly basic constituents of a primeval atmosphere, such as H<sub>2</sub>O, CO<sub>2</sub>, CH<sub>4</sub>, NH<sub>3</sub>, etc. We denote similar slight variants, or different relative proportions, of these molecules as a', b'... or a'', b'', etc. Compounds or polymerized forms from monomers are respectively denoted ab, abc; or aa, aaa etc. The molecule O<sub>2</sub>, which has a special significance in this discussion, we indicate by e. We denote organic molecules involving similar, relatively abundant, atoms by the upper case letters A, B; A', B' etc. We emphasize the highly schematic nature of this notation, intended only as suggestive regarding its chemistry, and adaptable for a generality of situations in potentially life-producing environments. In more detailed prebiotic reaction sequences numerous alternative components and branching pathways are considered. But a key idea, that we use below, is that of feedback in the interactions of mixed chemical systems.

The first stage of our scheme is an endothermic reaction sequence induced by insolation in the presence of a catalyst (when fully self-supporting) during the hours of daylight. It could involve some very simple proto-carbohydrate such as formaldehyde (H<sub>2</sub>CO) and subsequent formose-like reactions [43] or aldose condensations [44]. The essential result, significant to the present discussion, is a reasonably stabilized molecule, deriving from an oxygen reducing process, of enhanced CPE relative to its formative molecules in typical ambient conditions. In the terrestrial case, this product 'A', should be a simple monosaccharide—perhaps ribose (C<sub>5</sub>H<sub>10</sub>O<sub>5</sub>)—although it would then have to be within some suitably stabilizing molecular environment [45]. A more robust and simpler carbohydrate, known to form in abiotic conditions, is glyceraldehyde (C<sub>3</sub>H<sub>6</sub>O<sub>3</sub>). We expect the production pathway to be photosynthetic, probably, but not necessarily, along the lines considered also by Garrison et al. [46] (see also [47]). This 'A-subprocess' we write as:



The A molecule can be regarded as a (temporary) energy reserve for subsequent molecular interactions. We have indicated the key catalytic role of the second (conceptually) basic molecule B, but the brackets indicate that other molecules may also be involved in its action. This brings up the second stage of the process. Continuity of the A-B cycle is achieved during the hours of darkness, when the A type molecule is involved in a mildly exothermic (less energy released than was absorbed in the A process) reproduction of the B type molecule in the 'B-subprocess'. The B type molecule could be expected to have ring structure (as A), and, keeping in mind the amino basis of proteins, involve nitrogen. The appearance of bases related to pyrimidine (C<sub>4</sub>H<sub>4</sub>N<sub>2</sub>) in nucleic acids suggests this might play a prime role as a B-type molecule [48, 49, 50]. However, direct A-B synthesis along these lines in terrestrial conditions appears essentially ruled out [51]. More pointedly significant is the discovery [52] of a pathway to the nucleoside ribo-cytosine involving, as a key reactant, 2 amino-oxazole, (C<sub>3</sub>H<sub>4</sub>N<sub>2</sub>O). Figure 1 of Powner et al. [52] demonstrates this latter molecule in an A type subprocess, catalysing glyceraldehyde to the arabinose structure of a nucleosidic precursor molecule. Sugar + base assembly is then achieved, even if not by the more direct pathway supposed by earlier experimenters.

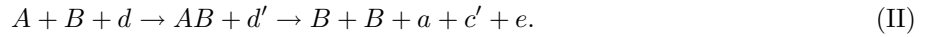
It appears likely that B would be in an energized state when performing its catalytic function, since candidate A type sugars have CPEs of typically several hundred (negative) kJ/mole, or eV per molecule. One kJ per mole is comparable to local thermal energies of atmospheric particles at ~ 20 °C, i.e. these molecules must have received significantly more than ambient thermal energies in their formation. At the peak of the

solar energy distribution photons have energies of about 2 eV, but Planck's distribution for the flux at shorter wavelengths, where the energy per photon rises inversely with that wavelength, entails a significant exponential decline by the corresponding wavelength ratio, impeding sugar formation in the absence of a catalyst. The work of Powner et al. [52] indicates a role for the greater binding energy phosphates being involved in intermediate or stabilizing reactions in the general terrestrial environment. Phosphorylated molecular bonds are well-known as an important vehicle for energy transfer throughout biochemistry. The relevant B type molecule, in the discussion of Powner et al., then looks to be phosphorylated 2-amino-oxazole. Powner et al. [52] also showed that UV irradiation of plausible former molecules is selective in leading to at least one stable AB form as a known nucleoside, noting the regenerative function of urea ( $\text{CH}_4\text{N}_2\text{O}$ ) in its assembly. However, the slow progression from less to more energized molecular structures requires continued cyclic reinforcement by an appropriate physical mechanism. Referring to the formalism of the previous section, we could expect to characterize the B-subprocess in the form

$$D\{A\} = k_1B + f_1(t), \quad (8)$$

where  $D\{A\}$  denotes the rate of sugar molecule formation, in dependence on the concentration of B type catalysts, but also with a dependence on the variable solar insolation, mainly through some environmental function (e.g. local temperature)  $f_1(t)$ . Equation (8) follows the asymptotic form of the Michaelis-Menten law, i.e. when the reaction has progressed to a uniform rate. This point is discussed in Appendix I where we consider the parallel treatment of [38] that does include the full Michaelis-Menten form. In practice, the coefficient  $k_1$  would be, to some extent, time-dependent through the Arrhenius relation, because the number of sufficiently energetic photons change during the day; however, to progress the present simplified analysis, we treat it as a constant (for a more detailed discussion cf. e.g. [53]). Within the timescales of operation of this initial sugar generation we expect other (inorganic) background factors (e.g. a, b, c type molecular densities) to remain fairly constant.

For the B subprocess we write:



In a similar way to equation (8), we then write

$$D\{B\} = k_2A - k_3\mathcal{A}, \quad (9)$$

where  $\mathcal{A}$  indicates the formation of the intermediate AB compound, for which we can write  $\mathcal{A} = k_4(A + B)$ . Note that equation (9) represents overall reactions; there could be intermediate steps, not directly indicated in equation (9).

If we now differentiate equation (8) again and substitute from (9), we obtain

$$D^2\{A\} = -\omega^2A - \lambda D\{A\} + f_2(t), \quad (10)$$

with  $\omega^2 = k_1(k_3k_4 - k_2)$ ,  $\lambda = k_3k_4$  and  $f_2(t)$  is the oscillatory function  $f_1' + k_3k_4f_1$ , together with other small contributions arising from time derivatives of the coefficients  $k_i$ . Equation (10) is of the same form as equation (3), which allows us to expect a consequent (slow) exponential growth in the appearance of sugar type molecules, if  $\alpha > \mu$ , as given by (5) and (6).

In a similar way we can also show that B satisfies the similar form

$$D^2\{B\} = -\omega^2B - \lambda D\{B\} + f_3(t), \quad (11)$$

with  $f_3 = (k_2 - k_3k_4)f_1$ . Since we expect that the oxygen molecule  $e$  to be liberated as a byproduct of the B subprocess, i.e. in a constant relationship to the number of B type molecules, it too should show a similar steady exponential growth and, by implication, the CPE of the process as a whole (again keeping in mind that equation (10) and equation (11) have a summary character regarding the complex of chemical interactions).

A key point is that the natural timescales ( $1/\omega$ ) for both A and B subprocesses should be of order half a day in typical terrestrial conditions. Such timescales, demonstrated by Weber [50] for feasible real molecules, and appearing also in the nucleoside assembly pathway of Powner et al. [52], are relevant to the susceptibility to diurnal resonance for the whole scheme. The AB compound has an important regulatory role that we consider further in what follows. The A-B process overall can become both multiplicative and CPE-aggrandizing, thus showing elementary lifelike properties. But the full range of possible formative A and B components include molecules that are not so easily formed naturally on Earth. It is feasible that the very first such molecules may have entered the atmosphere from space [54, 55]. Once here, continuation of the A-B process caused their continuous multiplication, increasing net CPE, and subsequent evolutionary development.

From the data given by [56], a given atmospheric molecule can be shown to experience  $\sim 10^{15}$  collisions during the hours of daylight. Later, we argue that a daily natural photosynthesis process could be expected to produce A type molecules at a proportion of up to  $\sim 10^{-12}$  of other constituents. If there is a single active B type molecule available, then there is a reasonable probability of an encounter with an A type in the lower atmospheric regions in a single day, resulting in the formation of an AB type precursor.

Summarizing, the A-B cycle acts as a parametric oscillator in a near-resonant condition with the diurnal rotation. During the day, B catalyses the production of more A molecules from initial constituents. During the night, A-type molecules condense towards the B type and, at some lower temperature, this leads via the intermediate AB stage to the regeneration of energized B molecules. Sometimes a somewhat different molecule  $B'$  may be favoured. The very wide variety of possible by-products of sugar-amino acid interactions is well-known. The possibility of rival catalysts B and  $B'$ , or control compounds AB or  $AB'$ , institutes a Darwinian evolutionary process on the basis of preferential selection in a given environment in relation to efficiency of the resultant resonance for surviving CPE generation, even at this fundamental level [57]. This simple AB or  $AB'$  dichotomy has an analogue in the 'black' or 'white' conceptualization of *Daisyworld* [58]. Early biospheres on planets with moderately uniform topology, could thus be expected to develop a coarse zonal structure.

If growth exceeds dissipation, the process continues to grow, but not simply in linear amplitude of excitation, or numbers of A and B molecules. The available modes of carbon-based molecular chemistry allow for the formation of more complicated, higher CPE, structures. In due course, B,  $B'$  etc. may become not just single molecules, but groups of more complex molecules, genes, or macrostructures, that co-operate towards the resonance-driven CPE enhancement. There should then be not just one A-B cycle, but a diversity of such cycles, filling in the available bandwidth for which  $\alpha > \mu$ .

Note that the compound AB molecules are reproduced as part of the cycle. Ambient AB types (protonucleosides) carry 'memory' of a higher insolation environment. Fewer new B molecules may emerge if the insolation increases, or more complete the process if the insolation declines. The presence of the AB compound thus contributes to homeostasis and tends to promote survival of the local protobiotic molecular ensemble through its negative feedback. We will compare this feedback concept with that of Pavlidis and Kauzmann [38] in the appendix. In due course, and with the steady increase of biospheric CPE, AB type molecules themselves compound to more complex  $ABAB'$  forms, made possible by the versatility of suitably energized bonding. However, maintenance of the driving resonance implies that the mean daily B molecule production must always

be maintained by the ensemble, implying, at a basic level, the demise of individual ABs.

ABAB' (etc.) molecules retain more specific information content about environmental variation, assisting multiplication and CPE aggrandisement, but in a more specifically defined environment (due to the implication that separate links in an ABAB' type molecule have separate, complementary regulating functions within a single chain). Part of the definition of such a local environment results from the distribution of A, B, AB, and ABAB' type molecules within it. The local feedback effect depends on the properties of these molecules, particularly regarding local environmental change. The slow build-up of more complex ABAB' structures also deposits a proportion of the CPE increase into fewer molecules, giving a proportionally large increase in the energization per molecule of longer protonucleotide chains. Surviving development, involving these more complex sequences, appears as controlled homeostatic behaviour, with highly energized long molecules imparting heirarchical structure into the control function. The local conditions for this increasing specification and control tend to differentiate the protobiosphere into *lacunae* containing small local centres of organization. Since the roles of air, water and sunlight are clearly implicit, near surface layers of the sea, including rock or tidal pools, may have provided an early natural realization of this scenario [58, 59, 60].

Given the near-homeostatic property required for growth from equation (6), reproduction of the particular molecular sequences involved in it ('replication') will be a natural characteristic. In other words, the restorative quality built into the dynamics means that other particle combinations than those originally energized will share the same local energization, provided they have the same, or very similar, properties. *A key question is then: at what stage does this 'bottom up' restorative development of the biosphere start to look more like 'top down' regulated replication of the lacunae?* Growing complexity of the latter seems likely to make the underlying biospheric driver increasingly less obvious as evolution proceeds; however, the fundamental role of biospherically generated global reagents, such as  $O_2$  or  $CO_2$ , shows that lacunal life can never be entirely self-organized. Local CPE excess should also assist the synthesis of new C type molecules (enzymes, protosomes, cytoplasm etc.), outside the catalytic ABAB' protonucleotide sequence, but maintained through subsequent multiplication if promoting survival of the underlying resonance. C type molecules are therefore peripheral, in the sense of Weismann [61], unless, perhaps, at some stage their functioning promotes the underlying resonance more effectively than the AB process itself, which would convolute the evolution. Something like this may have occurred with the transition from an RNA to a DNA based cellular organization. But, at a primary level, C type molecular structures are likely to evolve an increasing role in defining lacunal parameters and boundary conditions consistent with net homeostasis. C type molecules may also have an important effect in determining at least some of the constants  $k_i$  in the foregoing equations and therefore the 'tuning' of the system in response to the diurnal variation. Increasing molecular complexity, specific replication effects and control within lacunae, supported by enhanced CPE, thus become essential features of self-sustained development [62].

One single resonating A-B process can be conjectured to have survived as the predominant outcome of early lifelike development on Earth. This is suggested by the prevailing chiralities of evolved biotic molecules and the preference for one particular carbon isotope in living structures [19, 63]. This same A-B process might still be present within the ongoing biosphere, but the original molecular behaviour should by now have been considerably masked by secular evolution in a way parallel to the way the atmospheric composition has changed. The 'Goldilocks planet' scenario [64] is thus intimately related to the natural timescales of the A-B process linked to the energy variation of the diurnal cycle. After  $\sim 10^{12}$  such cycles, with a 'random walk' analogy to the general progress of evolution, it is tempting to expect lengths of surviving ABAB' type nucleotides to have reached  $\sim 10^6$  pairs. However, details of the relative numbers of the various organic molecules enhancing

local homeostasis, increasing CPE, reproducing participating molecules and maintaining the primary resonance become increasingly complex with the evolving biosphere, and are essentially a topic for future development. We offer further suggestions on the growth of complexity consistent with our model and compared with some observational data in Appendix 2. <sup>6</sup>

There may also be other periodicities in the response spectrum. The annual cycle, clearly reflected in the seasonal behaviour of advanced plants, is an obvious example. There is no comparable electromagnetic flux variation associated with the Moon, but a weak resonance with the lunar cycle appears to have affected the development of various life forms [65]. But such longer period components cannot have had the same basic driving function as the daily cycle, that reaches right down to abiotic photosynthesis.

We thus surmise that the original A subprocess was such a natural photosynthesis, as in the model of Garrison et al. [46]. In that case, equation (9) in mind, the mean daily yield of oxygen should be  $\kappa\rho\mathcal{F}A\tau m_0$ , where  $\kappa$  is the absorption coefficient,  $\rho$  is the density of sea-water,  $\mathcal{F}$  is the relevant ionizing solar photon flux integrated over an effective  $T$  seconds per day,  $A$  is the total projected oceanic area and  $\tau$  is an effective depth from which oxygen molecules of mass  $m_0$  can escape from the water surface. Some of these quantities can be given fairly definite values (for example,  $m_0 \approx 4.8 \times 10^{-26}$  kg), while, taking into account projection or mean attenuation factors, we can write  $T \sim 10^4$ ,  $A \sim 10^{14}$  (SI). The mean incidence of ionizing (pre-ozone-layer) photons can be estimated as  $\kappa\rho\mathcal{F} \sim 10^{20}$ . If we take  $\tau$  to be of the order of the mean free path of water molecules, so that a liberated oxygen atom within  $\sim 3 \times 10^{-9}$  m of the surface could escape, we will find a daily yield of  $\sim 10^6$  kg. The effective driving amplitude  $f_0$  in equation (5) thus appears of an appropriate order of magnitude ( $\sim 10^{-12}$ ) to give rise to parametric amplification. Oxygen from this process would not be expected to build up in the absence of life, however, due to its intrinsic chemical activity [66].

A comparable natural A subprocess is indicated by the diurnal pattern of hydrogen peroxide distribution on Mars [67].<sup>7</sup> Hydrogen peroxide is a well-known product of oxidative metabolism. In that case, however, the observed proportional amplitude of CPE excess ( $\sim 10^{-8}$ ) is some four orders of magnitude greater than would correspond to abiotic generation. None of the foregoing coefficients in the daily oxygen release rate would be significantly greater on Mars and some (e.g. the oceanic area  $A$ ) are considerably less. This anisotropy can thus be regarded as an accumulated protobiospheric response as in equation (6). The actual mean diurnal rate of CPE aggrandisement on Mars would then be  $\sim 10^{-8}$  that of the Earth, assuming the two planets are of about the same age.

The planet Jupiter is also of interest, not only because of the superficial resemblance between its zonal structure and Daisyworld models, or the possible detection of organic molecules in the atmosphere [68]. The Great Red Spot (GRS), and perhaps other comparable features, pose an obvious problem in relation to their longevity and dynamics. Close-up monitoring of the Jovian atmosphere shows that not only must the GRS be essentially dissimilar to other atmospheric cyclonic features in the belts, that follow formation and dissipation timescales comparable to expectable meteorological effects (i.e. a few rotation periods), but that it actively

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<sup>6</sup>From a teleological standpoint, since maintenance of the order of energized complex systems becomes their apparent purpose, interactions between separate such systems (highly developed ABAB' type forms) will become their preoccupation. Interactions between complex ordered systems may sometimes be statistically summarized, in their outcome distributions, by appropriate logistic equations, although these are too simple for the level of individual interactions. It is known that such equations permit 'evolutionarily stable' (ES) solutions, relating to observed numbers of surviving species. Coefficients in the relevant logistic equations reflect environmental and ABAB' type molecular fitness factors, but it must become increasingly difficult to derive, in a general way, what particular factors have resulted in the observed ES configurations of the Earth, or if indeed they have longterm stability.

<sup>7</sup>The abundance of H<sub>2</sub>O<sub>2</sub> on Mars was confidently determined by Encrenaz et al. [67], as distinct from the provisional identification of CH<sub>4</sub>.

propels itself against dissipative turbulence in its surrounding belt. The long-term survival of the GRS against this atmospheric turbulence can be estimated (from simple aerodynamic comparisons, e.g. with aircraft) to involve a power injection of order  $10^{16}$  W. The mean insolation over the GRS could be reckoned as of this order, or perhaps an order greater: so that the Sun is again able to drive the process energetically. The question of how this power might be transferred into the organized restoration of the GRS remains to be clarified, but it is clearly a candidate for lifelike behaviour in the context of our generalized model for primitive biospheres.

Having speculated on initial molecular possibilities, we can follow clock-like processes in cellular contexts of Earth's early biosphere that can be checked by observations, for example, with the prokaryotic cyanobacteria or perhaps even their photoautotrophic ancestors in Archean times [69]. Photoautotrophic species appear to be among the oldest, biologically accepted as living, inhabitants of the Earth. Microfossils of cyanobacterium-like microbes, discovered in well-preserved sedimentary and volcanic rocks of the Archean Eon, point to their presence as early as  $\sim 3.5$  billion years ago [69]. Some of these fossil species from Western Australia are physiologically compared to subsequent cyanobacteria of the genus *oscillatoria*. Biological evolution of the prokaryotic clock is considered to have paralleled the geological evolution of Earth [65]. If the primitive clock was the usually accepted ultradian oscillator, metabolic periods of early photoautotrophs should have been in synchrony with the phase and short-period rotation of the Earth [28]. For both organisms and Earth, the relevant periods are less than 21 h. It becomes an interesting question to research from this perspective, if the ultradian clock of today could have been yesterday's circadian clock.

In conventional biological terms, cyanobacteria that survived successive geological eras would have been those with biological clocks that best synchronized their physiological and metabolic processes with the prevailing environmental cycle. Within a population, those with the best clock, at the given era, should have an adaptive advantage, in the sense mentioned at the beginning of section 4, enabling them to outnumber or displace those with a clock that generally ran too fast or too slow. The adaptive value of this type of biological clock has been well demonstrated in the laboratory with various strains or mutants of cyanobacteria having different free-running periods [70, 71, 72]. Oxygen-producing cyanobacteria steadily changed the physical atmosphere of Earth from a reducing to an oxidizing environment. The level of UV in the insolation, at any era, contributes to the responsive and adaptive production of photolyases (repair mechanisms), photoreceptors, and circadian systems.

Given the fundamental role of the insolation in our argument, we should expect that as organisms evolved, a robust biological timing system enabling survival against particularly disturbing forms of radiation would have been conserved through to present-day species. The underlying problem, viewed in teleological terms, is optimization of the frequency range of insolation acceptance to maximize energy uptake yet minimize likely photon damage to controlling molecular structures. In this way, more sensitive processes, such as nucleic acid replication and cell division, should occur late in the day or at night. We may usefully compare, for our analysis, the biochemical model of Pavlidis & Kauzmann [38] for the circadian oscillation of photoautotrophs and Appendix I summarizes such a comparison.

## 6. Results and conclusions

Although many laboratories have pursued Miller type experiments, regenerative, evolving nucleic acid development *ab initio* has proved elusive. But, in the classical form [31]: (a) there were no 'seed' molecules entering the mixture from the interstellar medium. Now we know that the interstellar medium itself has

such molecules and occasionally they could enter the primitive mixture. This, in turn, relates to the ‘Cosmic Life’ scenario [[73], which stems from the basic difficulty of getting cellular life started, by chance, within the lifetime of the Earth. It is significant to notice that the initial starting and the subsequent growth in a suitable environment are quite different things; (b) the role of a resonance between molecular processes and a diurnal energy cycle was not considered directly.

This cycling need not, of course, be of just 24 hour period, because (1) the Earth’s rotation rate has varied and (2) the original A and B type molecules may have come from some other body (e.g. a comet) with a fairly different rotation period and surface conditions, while (3) experimentation is essentially about looking for key factors in the development of life. If a resonance effect plays some part, this could be tested over quite different timescales to check if there is a parametric growth effect, in a primitive molecular ensemble, at *any* period. Note that the growth in equation (6) does not require exact resonance with any particular single frequency. Oxygen production is a feature of our scheme and its general release into an atmosphere has been taken to measure the mean level of biospheric CPE enhancement. It would be useful to demonstrate this experimentally, keeping in mind that initial levels of such production should be extremely slight if the model is to be compared with terrestrial planets.

There is an implication, from Muller [39], that if there were such a resonant molecular cycling effect, it should be of longer structures, such as the ATP molecule. With smaller molecules, and terrestrial conditions of the present time, an initial A-B process may no longer be directly observed due to evolutionary development. But life emerging from prebiotic conditions is unlikely to begin with large polymers. An important purpose in experimentation is to define or check on possible a, b, c type candidates that lead to A, B type molecules [74]. Within the present context, we specifically ask: what natural timescales for relevant processes could be comparable to a 24 hour timescale? From our discussion in Section 5, this could more specifically direct itself to radical analysis of circadian rhythmicity. That presently observed circadian rhythmicity is controlled by modern genetic structures is not in doubt; but, regarding how this originated, we look for a deeper level of rhythmic organization, continuing from proto-biotic through to modern times.

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## Appendix 1: Comparison with the Pavlidis and Kauzmann (PK) model

The PK model is a 3 stage model for an established cell (lacuna, in the terminology of our ('ABC') model) operating homeostatically in a more or less constant environment, apart from the diurnal variation of sunlight: 1. During the hours of daylight monomolecular sugar (denote as X, and let be conceptually similar to the A-type molecules) is produced from supply chemicals (G) that are taken to diffuse into the lacuna at a more or less constant rate. 2. In the evening, X molecules are converted into catalyst activator molecules (Y, taken to be ADP in the Pavlidis and Kauzmann discussion; these may be regarded as equivalents to the B type molecules above). The production of the Y molecules is associated with a feedback action of the activated catalyst  $E_1^+$ , which returns to its deactivated state  $E_1$  at the completion of the process. (E types are C type molecules in the language of the ABC model.) 3. At night, the Y molecules disappear at a more or less constant rate, and the lacuna is returned to approximately its initial conditions by dawn of the next day. The removal of the Y molecules is associated with the activation of another catalyst  $E_2$ , which converts them into Z type molecules that diffuse out of the lacuna and back into the environment.

These 3 stages are represented, mathematically, by 3 equations that reduce, in asymptotic form, to

Stage 1

$$D(x) \sim g; \quad D(y) \sim 0; \quad y \sim 0, \quad (12)$$

Stage 3

$$D(y) \sim e_2; \quad D(x) \sim 0; \quad x \sim 0, \quad (13)$$

Stage 2

$$D(x + y) \sim 0; \quad y = \text{const.} - x \quad (14)$$

(where lower case letters scale with numbers of the corresponding upper case chemicals).

The constants that appear from the integration of the differential equations are adjusted so that the 3 stages line up, more or less, as the sides of a triangle in the  $x, y$  plane, and locate this triangle so that its horizontal and vertical sides are close to the  $x$  and  $y$  axes, respectively, while both  $x$  and  $y$  remain positive throughout the cycle. The original PK equations are actually more complicated than those given above, where the chemical reactions are set up so as to follow the Michaelis-Menten law, and they introduce an appropriate function ( $v$ ) that switches the metabolism from one stage to another, but this function only decisively affects the (small) process-change regions. The function  $v$  takes the standard Michaelis-Menten form  $v = v_{\max}s/(k + s)$ , where  $s$  represents a (time-dependent) concentration. The rate-affecting function  $v$  then tends to a maximum as  $s$  tends to its largest steady value, and will be asymptotically close to this value for settled (non-initial) values of  $s$ . Since the key processes depend on the external photon flux, rather than local variables, we assume that the asymptotic forms adopted in the main paper will be relevant to describe the mean behaviour of the system.

The energy of stage 1 is understood to come from the environment, while that of stage 2 (activation of the enzyme) is associated with the solar photon energy (the solar maximum flux equates to several thousand blue-green photon eV per second per square micron). The energy of the third process may be taken to derive from the excess energy of the Y type (ADP) molecules, which would be sufficient to allow the cell to remain homeostatic in a somewhat variable and dissipative environment. (The excess energy of the diphosphate bond is about 0.34 eV.)

Pavlidis and Kauzmann recognize the central role of feedback in their oscillator model, and also that this should be negative to ensure stability or quasi-homeostasis. A primary requirement, to allow their model to be compared with real cyanobacteriae, is that of temperature independence of the cycle's period. In the PK model this is achieved by a temperature-dependent detracting of the main activated enzyme  $E_1^+$ , so that any possible increase of activator rate with temperature is compensated by a lower concentration of operand (thus evolved). In the AB process, it is increased accretion of the B-molecule in the AB compound that inhibits its free catalytic function.

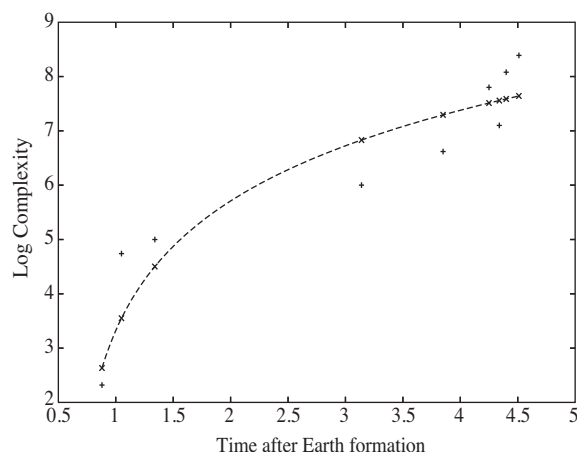
Concerning the PK cell, although there is homeostasis, there is a slight change for the biosphere as a whole, associated with the steady replacement of G by Z type molecules. The removal of excess energy through the Z type particles thus implies a steady increase of external biospheric energization. Alternatively, the powering of additional enzymes, such as  $E_2$ , demonstrates the role of primary oscillatory energization applied to cell development.

In an origin-of-life situation, there are no cells and no separate enzymes. There is, however, a sugar supply of some sort (G) a daily insolation and the potential biosphere. It is also known that elementary RNA shows an equivalent autocatalytic function ([75, 76]). The very high natural concentration of virus-like particles in typical near-surface marine environments (cf. e.g. [77]) is suggestive in this regard. We may then compare the PK equations to the ABC (biospheric) model as (1)  $D\{x\} = -k_3e_1 + g(t)$  (compare with equation (8) above), and (2)  $D\{y\} = -k_7e_2 + k_3e_1(xy)$  (compare with equation (9)). If  $y(\equiv B) = \text{const.} - ke_1$ , and there is a further connection between  $e_2$  and  $x$  (not given by PK) then these modelling equations are of essentially similar form.

## Appendix 2: The Development of Complexity: Simple Model and Comparison with Data

In Section 5 we visualized a slow build-up of more complex ABAB' (etc.) structures occurring within local lacunae. Such ABAB' structures reflect a more specific information content on environmental variation; the different links in an ABAB' type molecule having complementary regulating actions. This, in turn, relates to the feedback affecting the distribution of A, B, AB, ABAB' (etc.) type molecules within a lacuna. This may be understood as a pyramidal or 'sandpile' type arrangement (cf. [78]), with a characteristic slope  $\gamma$  coming from the stability requirement that  $\delta U/\delta E\gamma$ , where  $\delta U$  denotes a mean fluctuation of local energy density (probably usually associated with temperature changes over the timescale of a day) and  $\delta E$  corresponds to the typical inter-link binding energy of an ABAB' type molecule. We could then expect numbers of corresponding AB, ABAB', and ABAB'AB'' (etc.) type molecules to decrease in proportion to  $\gamma$ , while the overall height of the pyramid (i.e. length of the longest molecule in the pile) increases in proportion to the general growth parameter  $\alpha - \mu$ .

Let us further suppose, in agreement with the observed numbers of fundamental base (B-molecule) types, that there is some small number  $n$  of these, though the number of such linked (AB) pairs on a molecular chain  $N$  can become indefinitely large. We might further consider genetic 'word' lengths, consisting of some other small number,  $m$  say, of the  $n$  types on the long chain, to be used to denote specific elements of control information, but we can ignore this refinement for present exploratory purposes. The location of a particular controlling element on the chain is likely to be significant in relation to its action, since the diffusion time for different interactions relates to the geometry of the overall arrangement. We can therefore consider the number



**Figure 1.** The growth of complexity, plotted as  $\log \Psi$  versus time measured in By after formation of the Earth, following [73, Figure 5.7], is here modelled by a simple permutation of B-type molecules on a developing molecular chain that grows from the underlying driving mechanism acting within a lacunal hierarchy.

of permutations of the  $n$  AB complexes on the chain of  $N$  as

$$P_n^N = \frac{N!}{(N-n)!}, \quad (15)$$

and let us define a ‘complexity’ measure  $\Psi$  as  $\log P_n^N$ . Since  $N$  can become quite large, particularly by the time such a complexity reaches the scale that we could compare with recognized terrestrial biological species [73], we can use Stirling’s approximation to write  $\Psi \approx n \log N + n$ . This can be used as the basis of a fitting function ( $\Psi_t$ ) to apply to observed complexity values and we show an example of this in Figure 1.

In matching general considerations to the terrestrial case, it may be asked: why is there only one A type, just a handful of B types, or a score or so of basic C type molecules (on Earth)? The answer could be expected to involve specifics of the energy yield optimization derived ultimately from the A molecule’s temporary excess. This process has to meet stringent resonance requirements in order to survive and evolve successfully, and the number of primary catalytic inter-reactants satisfying the natural timescale condition (B types) is probably small. In subsequent refinements of the de-energization, lower energy differentials are involved, and many more available states, corresponding to the C types, may be expected [79]. In any case, the effective value of  $n$  for the alternative control components is small relative to  $N$ . In our model, from equation (4) above,  $N$  should rise exponentially with time within the Stirling argument  $n \log N + n$ , so that if  $N$  varies like  $e^{a_1 t + a_4}$ , say, where  $a_1$  and  $a_4$  are parameters, we can write

$$\log \Psi_t = a_2 \log(a_1 t + a_4) + a_3. \quad (16)$$

The logarithmic character of the growth of  $\log \Psi_t$  in Figure 1 can be recognized from its shape, which confirms, since  $\Psi$  is itself a logarithm, that the general growth of the number of AB type molecules must exponentiate. The position of the logarithmic singularity near the origin is essentially fixed by lowest complexity value in our data, namely that of viruses ( $\sim 200$ ) at a time  $t_0$  about 0.9 By after the formation of the Earth. Hence,  $a_1$  and  $a_4$  satisfy  $a_1 t_0 = a_4 + \epsilon$ , where  $\epsilon$  is some small positive number. The characteristic e-folding

timescale ( $a_1$ ) of the example shown is  $8 \times 10^{-13} \text{ d}^{-1}$ , which is lower than the growth parameter derived in Section 4 of the paper above, but the growth here refers only to the peak length of control molecules in the lacunae. The relationship of this peak to overall molecular development in the biosphere depends on the ‘geometry’ ( $\gamma$ ) of the growth pyramid mentioned above. The scaling parameter  $a_2$  fixes the amplitude of the growth in complexity; that is, by 5–6 orders of magnitude over  $\sim 4 \text{ By}$  since the appearance of viruses. This parameter too ( $a_2$ ) could be expected to be not entirely independent of  $a_1$ , but satisfy some relationship involving  $\gamma$ ; such as the relationship between  $N$  and the overall number of lacunal molecules. The remaining parameter,  $a_3$ , fixes the location of the curve in relation to the ordinate axis. The data was precalibrated according to a given scale of molecular ‘wording’. The complexity of a simple virus, the number of ways in which a basic coding unit could be arranged within the number of available places, then turned out as about 200. Because the  $y$ -axis is logarithmic here, this scaling simply takes on the role of an additive constant.

The treatment presented here is indicative and exploratory only. Delsemme [73] discusses reasons why potentially useful control information tends to be increasingly lost, or disregarded, within cellular genetic function. Other factors that must come into play include the faster than average early rotation speed of the Earth, greater former proximity of the Moon, together with other general vagaries of the terrestrial environment. Such external variations occur as well as the possibility of significant convolutions in the way the controlling feedback that enhances overall yield of the basic resonance may work, for example with the switch from RNA to DNA control structures, or more sophisticated ‘intelligence’ (cerebral or electronic) that operates on greater amounts of information at higher levels.