The Primordial Bubble:
Water, Symmetry Breaking, and the Origin of Life

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18.1 A PRELUDE ON THE SYMMETRY OF ORIGINS AND THE INTERFACIAL NATURE OF SELF-ORGANIZATION

Origin problems are the most conjectural and qualitative of scientific questions. It is hardly surprising then that the origin of life, like the origin of the universe, lacks uniquely defining quantitative assumptions and initial conditions. Individually, they are Fermi questions of a functional sort, requiring the use of a first-principles conceptual approach to a situation of fundamental quantitative ignorance. Indeed, concepts useful in the exploration of one question significantly help our investigation of the other: this is true for underlying the cosmochemistry of life is a cosmophysics hinting at congruences of symmetry-breaking problems in both the origin of the universe and the origin of life. But where theorists such as Lee Smolin have sought to apply the ideas and concepts
of evolution to cosmology (Smolin, 1997), in the present work we present a new concept of physical self-organization and apply it to (prebiotic chemical) evolution.*

The physicist’s tool of symmetries and their breaking is applied quite naturally here, as issues of symmetry at the grandest scale are fundamental to the Anthropic Principle. This is because the Anthropic Principle can be elegantly read as a statement of symmetry breaking with regard to the “Symmetry of Probabilities” (which is itself the basis of the physical worldviews of Copernicus, Newton, and Einstein—e.g., “There does not exist a privileged position in phase space”). Hence, the numerical tuning of cosmological constants is a qualitatively similar problem to that of the biochemical tuning of life: How did each arise out of all the possibilities and how was this “symmetry of probabilities” broken?

We emphasize in the present work the critical role of microenvironments and their interfaces in the act of symmetry breaking and the consequent building of structure. We suggest that at every physical scale it is the boundaries of the relevant microenvironments that play an essential organizing role. Generalizing this idea further naturally incorporates the notion of symmetry breaking. In particular, we propose the following: that least-action and least-energy principles applied to symmetry breaking leads to the universal role of interfaces as the symmetry-breaking microenvironments necessary for self-organization to occur. The key points† of this thesis are arranged alliteratively as follows:

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**Lerman’s Lemma of Self-Organizing Systems**

Self-organization over all scales of nature occurs in dimensionally nested hierarchies of “first-order”§ microenvironments; and where the matching of the space and time scales of the microenvironments to the self-organizing process is critical.

**Lerman’s Law of Self-Organization**

Self-organizing microenvironments are found specifically at the interfaces between phases, regimes, gradients, forces, and/or dynamic processes.

By microenvironments, one means the (heterogeneous) subsets of a bulk-average environment, whatever the scale. To understand what is meant by a “nested hierarchy,” one can think of nested Russian dolls. Taking the terrestrial ocean as example, microenvironments can range from the sea-surface microlayer to the region surrounding a single bubble. These two microenvironments are, in fact, intimately related; a near-surface bubble being a microcosm of the air-sea interface, which itself brings into being the unique chemistry of the sea-surface microlayer (MacIntyre, 1974a, 1974c; Sieburth, 1983). Hence, an example of two elements of a nested hierarchy of microenvironments

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* Because of the highly interdisciplinary nature of these problems, this paper uses ideas from a variety of fields. We start with a bit of abstract conceptual physics in this section, evolve by Section 18.3 to the functional requirements of prebiotic chemistry, and starting in Section 18.5 devote the majority of this paper to explorations of planetary geochemistry and the origin of life.

† This work is part of an effort to develop a more general approach to self-organizing processes. See Lerman (2002d) on a new form of symmetry breaking in relativistic nuclear collisions, and Lerman (2003, 2005c) for initial presentations of the above phenomenologically based laws.

§ By first-order microenvironment we mean most probable. It need not be a first-order phase transition, although the boundaries of these regions will often involve a phase-transition of the first-order.
that, as we will clearly demonstrate in this paper, have a set of chemistries and chemical physics unique and highly differentiated* from that of the ocean as a whole.

A simple illustration of self-organization that occurs at a boundary between dynamic processes would be “salt fingers” formed at the interface between opposing temperature and salinity gradients. The direction of a gradient-induced transport is essential, for a conservative quantity transported down its own gradient results instead in a mixed state of higher entropy. Indeed, driven by symmetry-breaking negentropic processes, self-organization at any scale must occur at interfaces almost by definition. The basic logic for this abstraction is as follows:

1) Symmetry and structure are inversely correlated.
   To illustrate this nonintuitive concept: Although a snowflake has greater structure, the water in a snowflake dispersed throughout a room as a vapor has much greater symmetry (the few reflection and rotation symmetries of the snowflake are dominated by the exchange symmetries of a gas).

2) Symmetry breaking → structure building
   By definition, the breaking of a symmetry changes the homogeneity of the property represented by the invariance (under the group operator) that describes the initial symmetry. The initial homogeneous state becomes heterogeneous, and almost by definition, structure is created. Structure building is the essence of a self-organizing process.

3) Symmetry breaking → boundaries
   Along with the creation of structure, a transition region (i.e., a boundary) between the now heterogeneous regions is created. Again, by definition, it is at the boundary (the interface) between these heterogeneous regimes that the describing symmetries are globally discontinuous, i.e., broken.

4) Boundaries and least energy → microenvironments
   Least-energy constraints lead to boundary conditions that, in turn, create geometrical and topological constraints on the now heterogeneous regions. At the scale of the symmetry breaking, these topologically constrained heterogeneous regions are the microenvironments that make up the structure.

5) Broken symmetries build upon (previously) broken symmetries
   By induction, the next level and scale of structure building will occur at these boundaries. One of the primary reasons for this is the decrease in entropic possibilities (in the relevant phase space) that necessarily exist at these interfaces.

6) Applying the above: Self-Organization occurs at the boundaries of microenvironments.

The logic described above derives from the fundamental fact that entropy, being a quantitative measure of disorder, is equal to the number of accessible states that look the same. In other words, entropy is a measure of the symmetries of a system; so breaking symmetries reduces entropy and creates structure. Moreover, Noether’s theorem (Noether, 1918) states that symmetries of a system correspond with conserved quantities; but from non-equilibrium thermodynamics we know that structure comes from systems that are far from equilibrium. This means that they involve systems in which some of the thermodynamic variables are not conserved, i.e., symmetries are broken.

It is hoped that these principles will have a philosophical component analogous to the way in which the variational principle embodies natural philosophy: and that the underlying logic and approach can be formalized and applied to many scales of phenomena. In addition, they are intended to provide critical insights into how and where (self-)organization takes place from one level of structure to the next. For example, an important consequence for astrobiology is that chemistries

* With respect to the world of chemistry and chemists, “This is distinct from a traditional chemical view, where heterogeneity is clear at the “beaker scale” (things are in the beaker or not) and at the molecular scale, but at scales in between, the chemist hopes to have an averaged environment.” Steve Benner (private communication, 2005).
based on different values of the fundamental physical constants, or “merely” different chemistries than our own water-soluble organics (Benner, 2004), will require analogous forms of microenvironments and their boundaries.

Alternative chemistries based on alternative physics, if they are to support the conversion of inanimate matter to structured living systems, will need to be evaluated not just with respect to bulk-state reactions but by their ability to form interfaces and microenvironments. This will restrict the number and nature of otherwise “plausible” variant chemistries (Benner, 2004). That is, if the alternative chemistries are to support the emergence of life, then a mutual incompatibility of two liquid phases would seem to be necessary. Attempts to use nonpolar solvents (such as methane on Titan) will be especially affected by this requirement. Without some form of polarity it is going to be tough going, for the polar nature of the underlying molecules is itself the symmetry-breaking template upon which the rest of the (self-)organizing process is built.

Certainly with respect to the phenomenological model described in this paper, the linkage between water, its symmetry-breaking interfaces, and its ability to support chemical self-organization in a prebiotic context is not just intimate, it is fundamental.

18.2 THE SCIENTIFIC PROBLEM OF PREBIOTIC CHEMICAL EVOLUTION

At every scale of the observable universe, the evolution from simple to complex is congruent with what we see. Thus, in this paper, we assume evolutionary self-organization regardless of the scale of the physics, chemistry, or cosmic geography. Hence, the origin of life is assumed to have occurred through the processes of prebiotic chemical evolution, presupposing successive generations of increasingly complex organic molecules combinatorially synthesized from earlier generations. Less obvious is how this combinatorial chemistry occurred or how the overall process of chemical self-organization was functionally supported at each stage of its occurrence in the prebiotic environment.

This latter point, the functional support of prebiotic chemical evolution within realistic planetary environments, is a requirement for any nook or cranny of a universe harboring ambitions to support the development of life, in any chemistry or form. It is a goal of this work to explore the mechanisms by which water, through its ability to form microenvironments, offers a fundamental functional support for the processes of prebiotic chemical self-organization.

Unfortunately, we know far more about the evolution of life on Earth, its false steps, and its successful building blocks, than we know about how it came about in the first place. But just as the biochemistry of contemporary organisms can be viewed as a “fossil” record of biogenesis, the geochemical physics of the contemporary Earth can help delineate the self-organizing processes underlying prebiotic chemistry on an early Earth, Mars, or any other terrestrial-like body. Taking the minimalist assumption of microscopic stochastic processes, we first consider the system and molecular-level requirements necessary to support a bootstrapping of molecular organization. Especially when considering alternative chemistries to the origin of life, it is critical to look at functional requirements from the standpoint of first principles, utilizing only the most basic and transparent of assumptions and imposed boundary conditions.

18.3 CHEMICAL EVOLUTION AS PROCESS: SYSTEM AND MOLECULAR LEVEL REQUIREMENTS

Continuing on in a Fermi question-like manner, we necessarily assume ignorance of the totality of reactions involved in the creation of successive generations of increasingly complex prebiotic organic molecules. But using the stochastic nature of physics applied to the simple-to-complex nature of chemical self-organization means that each level of self-organization evolved from broadly ranging consecutive chemical trial and errors. Some pathways will work, leading to products useful
for the construction of the next level of organization, but most will not. Hence, in the abstract, our
presupposed processes of chemical evolution require the existence of a global chemical engineering
system (Lerman, 1986, 1992, 1994a, 1994b, 1996, 2002b; Lerman and Teng, 2004a), which at
the system level amplify the probabilities of stochastic self-organization at the molecular level. For
this amplification to be effective at the system level, these still-to-be discovered processes must be:

- Probable (“had-to-have-been” mechanisms)
- Robust (highly efficient and rapid processes)
- Diverse (exploring many different possible chemical routes and mechanisms)
- Selective (for the good stuff)
- Semi-closed (able to retain useful materials in the total system)

At the molecular level, the race between the self-organization of increasingly complex organic
structure and its dissipation due to entropy requires the following functional operations:

- Selective concentration of the desired compounds needed as reactants
- Stabilization and coordination of these reactants
- Controlled energy and “directed” synthesis
- Cycle continuity (where the products become in turn the reactants for the next stage of the
cycle)
- And most importantly, in our least-assumptions approach, all must occur in a plausible,
indeed likely, geophysical/geochemical environment

An additional boundary condition for the chemical evolution of water-based carbon chemis-
tries is the existence of (micro-)environments promoting both heterogeneous and dehydration reac-
tions. This is because the polymerization of biomolecules almost universally\(^*\) requires an H\(^+\) and an
OH\(^-\) to be discarded, one each from either of the two sides of the forming bond. It is thermody-
namically very difficult for this expulsion of H\(^+\)OH\(^-\) (i.e., H\(_2\)O) to occur in an aqueous environment;
therefore, pure solvent (homogenous) chemistry in water seems unlikely to have been the primary
process by which organic chemistry bootstrapped itself up the ladder of complexity.

Contemplation of the above-stated requirements reveals that each embodies a form of symmetry
breaking in space, time, and/or thermodynamic variables. This is hardly surprising as each is an
organizational (anti-entropic) step leading to some local enhanced state of structure. One of the
goals of prebiotic planetary chemistry is the need to demonstrate the existence of a global chemical
engineering system that has all of the above functions, synergizes each to the other, and that had to
have been in existence on the early Earth or Mars. Our question of the origin of life now becomes:
“On what larger organizing processes can chemical evolution piggyback itself?” Or, in the parlance
of the previously stated “lemma” of self-organization: “At the scale of chemical self-organization,
what ‘first-order’ microenvironments exist and mutually connect with each other to support increas-
ingly complex cycles of prebiotic chemistry?”

Such a first-order system exists on the contemporary Earth: it is the bubble-aerosol-droplet cycle.
Described in Section 18.5, it is the most fundamental, robust, and far-reaching of geophysical/chem-
ical supercycles\(^†\) involving organics in and between the ocean and atmosphere. Comparing these
present-day terrestrial processes against the functional requirements listed above leads directly to
the bubble-aerosol-droplet model,\(^‡\) a phenomenologically based model with the potential to sup-
port chemical evolution in all its stages (Lerman, 1986, 1992; Lerman and Teng, 2004a). These

\(^*\) There are a very few biopolymers that do not, e.g., terpenoids like vitamin A.
\(^†\) It is a supercycle because of the many large complex hydrological cycles that are subsets of the whole.
\(^‡\) It has also been referred to by a variety of other names in the scientific literature and the popular press, including the
Bubble Hypothesis, Lerman’s Bubble Model, the Bubblesol Hypothesis, etc.
range from raw organic synthesis on the prebiotic Earth to the possibility of helping coordinate the transition from organic chemistry to biochemistry. Indeed, this model seems relevant to any terrestrial-like body with liquid water and simple organics. (In Section 18.15, we apply the bubble-aerosol-droplet model to an early warmer wetter Mars and consider implications for currently observed Martian geology.)

Over the many physical scales and modalities of our model, one common theme will clearly be seen: the organizing ability of the water-air interface, with these organizing properties themselves a consequence of the symmetry-breaking properties of water’s heterogeneous two-phase interface.

### 18.4 THE AIR-WATER INTERFACE AND ADSORPTION

The keystone elements to all of the above are the symmetry-breaking surfaces of the microenvironments of the air-water interface: from the surface of a freestanding body of water (static or flowing) to whitecap-induced bubbles and their aerosol progeny. The shape of the surface of each of these microenvironments is itself a consequence of least-energy considerations, as is the topologically closed nature of the bubble or its inverse droplet.

It is for this reason that one can look at bubbles and aerosols as complementary: one is a metastabilized fluctuation of air in water, the other of water in air. The continuous layers of bubble clouds formed beneath breaking waves are the analogs of low stratus or stratocumulus clouds in the Earth’s atmosphere (Kraus and Businger, 1994). Whether floating under water, bobbing at the sea surface, or drifting in the atmosphere it is the adsorption of amphiphiles that drives these self-organizing phenomena by decreasing the local surface energy and metastabilizing local fluctuations into microenvironments with organized structures capable of further organizing organics, metals, and larger scale particulate matter. Additionally, the dimensional projection from three-space to two-dimensional surfaces significantly decreases the entropic possibilities of molecular orientations and dynamics.

Central to the function and partially closed nature of the bubble-aerosol-droplet cycle is the ubiquitous bubble itself. Isolated bubbles come into existence as regimes of a gas in a liquid in which the surface of the bubble-water interface exists due to a local increase of surface free energy supported by the pressure of the vapor contained within the bubble regime. The surface energy per unit area is also termed the surface tension, but the latter is a potentially misleading term. Molecules on a bubble surface are not necessarily stretched under tension to form the surface; the term surface energy more accurately reflects the work required to bring molecules from the vapor phase (in the bubble’s interior) onto the bubble surface (Adamson, 1997). Hence, a potential energy difference exists between the surface and interior of a bubble, allowing well-matched surfactants the opportunity to build more complex and stable structures through the lowering of this energy. Adsorption is thus driven by a reduction of the surface free energy as material is added. Lacking the stabilization due to adsorbed materials, a pure water bubble without any sort of a skin has a surface lifetime of less than a second. With stabilization due to adsorbed materials (organics and metal ions), the lifetime in the open terrestrial ocean can range from seconds to hours.

Adsorption is highly selective and is dependent on the details of the system, which include the following:

- The charge configurations of the molecules involved, especially with respect to hydrophobic and hydrophilic components on a single molecule (i.e., surface activity)
- The ability of the adsorbed materials to couple to previously adsorbed surfactants
- The relative sizes of the bubbles and objects to be adsorbed
- The composition of the bubble vapor, materials to be adsorbed, and various impurities

The air-water interface is an organizational foundation relatively independent of the specific chemistry of the atmosphere and ocean. Also being a function of the (curved) air-water interface, this relative independence is likewise true for the aerosol-droplet (inverted bubble) phase of the supercycle.
The robustness of the selective concentration abilities of bubbles and aerosols is underlined by the large number of industrial processes (e.g., mineral flotation and secondary and tertiary petroleum extraction) based on the use of bubbles, foams, and aerosol processes (Lemlich, 1972).

18.5 THE BUBBLE-AEROSOL-DROPLET SUPERCYCLE: A UNIVERSAL PLANETARY HYDROLOGY CYCLE

On the contemporary Earth the bubble–aerosol-droplet cycle (also known as the bubblesol cycle) includes bubble formation and the adsorption of surface-active materials, bubble dissolution, and the non-equilibrium dynamics of bubble bursting (Figure 18.1). This leads to the formation of aerosols and their subsequent roles in atmospheric condensation, which in turn couples back to the bubble phase of this “supercycle” (Lerman, 1992).

**FIGURE 18.1** The Bubble-Aerosol-Droplet Cycle. (See color insert following page XXX.) There is every reason to believe that this cycle also existed on the early Earth and possibly Mars.
Remarkably, this complex supercycle is a process that requires only the disturbance of a water-air interface metastabilized by simple amphiphilic compounds. The rest follows from the fundamentals of chemical physics, being relatively independent of specific chemistry. Specifically, bubble formation is a physicochemical process due to the Rayleigh-Taylor instability (Sharp, 1984), its initiation requiring only the existence of an air-water interface disrupted by mechanical turbulent energy (from waves, meteorites, or geophysically active regions). Its maintenance requires only a metastabilization by simple amphiphilic compounds that would have come from meteorites (carbonaceous chondrites) and comets. This latter statement is strongly supported by the fact that 70% of the organic compounds in the Murchison meteorite are polar (Cronin and Chang, 1993). Hence, it is difficult to imagine something similar to the bubble-aerosol-droplet cycle not being active in both the early Archaean on Earth and a wet Noachian on Mars.

There are two bursting mechanisms possible for an isolated bubble (MacIntyre, 1972), each leading to the formation of a different class of particulate matter injected into the atmosphere (Figure 18.2). For small bubbles (<0.5 mm), surface tension-driven "jet drops" are formed, which in turn are the principal source of sea-salt aerosols. By some estimates this is the largest source (by mass) of particulate matter injected into the atmosphere. For larger bubbles (>0.5 mm), an instability mechanism due to the gravity-driven draining of the bubble cap leads to the formation of large numbers of "film cap drops," particles that are much smaller than jet drops. This mechanism is possibly the largest source (by number) of particles injected into the atmosphere, and may be the major source of cloud condensation nuclei as well.

Particles injected into the atmosphere by bursting bubbles have different airborne lifetimes depending on their mass and composition. The heaviest particles injected immediately fall back to the water, forming more bubbles upon their impact. Particles swept up into the atmosphere will enter into the complex tropospheric aerosol cycle (Figure 18.3). Some will act as condensation nuclei for precipitation, and some will be scavenged by other precipitation bodies. The condensation nucleus

![Figure 18.2 Formation of Jet Drops and Film Cap Drops from a Bursting Bubble. (See color insert following page XXX.)](image)

(a) For small bubbles (<0.5 mm) surface tension-driven 'jet drops' are formed. The material making up successive jet drops from a single bursting bubble comes from successively micromted layers of its air-water interface (MacIntyre, 1974c). (b) For larger bubbles a very large number of 'film cap drops' are produced by a gravitationally-induced instability mediated through the bubble cap's draining. (Photographs by Prof. Andrew Davidhazy, 2009. With permission.)
of an aerosol can undergo a number of hydration-dehydration cycles before eventually falling to the ocean surface. While in the atmosphere, these objects will be exposed to a variety of energy sources including solar radiation and the plasma and shock effects associated with lightning.

18.6 THE BUBBLE-AEROSOL-DROPLET SUPERCYCLE: A UNIVERSAL “ORGANIC WEATHER” CYCLE

Accompanying the water-air cycles are “organic weather cycles,” which seem to support each of the functional requirements for chemical evolution listed in Section 18.2 at both the system and molecular levels. This is because the very existence of this first-order cycle, its nodes, and processes implicitly depends on these same functional requirements. Once a bubble forms it tends to collect surface-active surfactants, this being the first of several different concentrating phenomena associated with bubbles, aerosols, and droplets. These concentrated materials, which are predominantly organic, stabilize the bubble allowing it to follow one of several pathways (Blanchard, 1975, 1983, 1989; Blanchard and Woodcock, 1957). Each of the pathways as described below offers additional concentrating opportunities. Dovetailing between steps moreover suggests that once organic matter enters into this cycle, it tends to remain within it; at each step being joined by freshly accumulated organics, ions, and heterogeneous catalytic surfaces.

Based on the Rayleigh-Taylor instability, even short timescale disturbances of surface water (lakes, seas, or intermittent turbulent flows) can initiate this cycle, which by analogy to the current terrestrial ocean includes the following:

2. Organic materials and selected metals, as well as clay particles, are preferentially adsorbed onto the surface of the bubble. A bubble being a micro-version of the air-sea interface, it will adsorb and become stabilized by those dissolved materials that congregate at the air-sea interface. These include fatty acids, alcohols, proteins, and polysaccharides as primary adsorbents; these organics, in turn, adsorb metal ions, colloidal silica, and clays (kaolin and montmorillonite) (Johnson and Cooke, 1980; Lemlich, 1972).

3. This stabilizes the bubble, thus leading to a highly concentrated resultant particulate, as the now organically “dirty” bubble dissolves or bursts.

4. A dissolving bubble yields an organic-rich residue, which can then nucleate other bubble formation, or be adsorbed in turn by other bubbles.

5. All of the above leads to a significant enhancement (up to a million-fold or better compared to bulk water) of organics and metals at the millimeter-thick microlayer of the air-sea interface (MacIntyre, 1974c; Duce and Hoffman, 1976; Sieburth, 1983).

6. The bursting of bubbles injects into the atmosphere particulate matter also rich in minerals and organics, the latter being as much as 40–60% of the mass (O’Dowd and de Leeuw, 2007). See Figure 18.4. Besides organics, concentrated up to a million-fold, these processes can yield up to a 10,000-fold enhancement in phosphates (MacIntyre and Winchester, 1969) and other scarce minerals and ions (MacIntyre, 1970, 1974a, 1974b).

7. The bubble-bursting process also provides a set of energy-producing possibilities capable of driving highly non-equilibrium chemical synthesis (see Section 18.10).

8. These injected materials are then coupled to aerosol formation and the subsequent nucleation of atmospheric condensation, leading to the further heterogeneous chemistry and non-equilibrium physics associated with rain and snow (discussed in Section 18.11).

9. Precipitation concentrates organics and minerals from the atmosphere. During this scavenging process, there is evidence for the formation of an organic skin around the precipitation object (rain, snow, hail) (Gill et al., 1983). Fog particles have been found to contain mostly organic carbonates, esters, and proteins, whereas the fatty acids in aerosols have been shown to have an oceanic source. Organics and transition metals from meteoritic sources are also scavenged and concentrated (Murphy et al., 1998).

10. These take part in a variety of heterogeneous physicochemical reactions including successive hydration-dehydration reactions that may be critical in the transition of organic chemistry to biochemistry (Section 18.12).

11. Atmospheric condensation closes this supercycle since both falling raindrops and snow directly deposit their organometallic chemistry on the ocean surface, efficiently inducing new bubble formation. Precipitation-induced bubbles are an important link in merging the oceanic and atmospheric subcycles of the entire bubble-aerosol-droplet supercycle, whereas wind-induced bubbles will tend to collect the rest of the material congregating at a sea surface.

12. Once organic matter, inorganic ions, and terrestrial and meteoritic particles enter anywhere into the bubblesol cycle, they tend to be continuously recycled and are therefore available for the self-organizing chemical-physical processes discussed in the remainder of this paper.

Throughout this supercycle, coupled hydration-dehydration cycles are abundant. As indicated above, this is of unique importance to the self-organizing formation of biopolymers, for essentially all biopolymers are formed through linkages derived from a dehydration reaction. Chang and Lahav (1982) have additionally shown that such polymerizing condensation reactions are greatly enhanced if the organics undergo hydration-dehydration cycles on salt and mineral surfaces. Fortunately for our intended prebiotic purposes, such organically rich hydration-dehydration cycles naturally occur at every step—e.g., at the concentrating sites at each phase of the bubble-aerosol-droplet cycle (bubble bursting, aerosol formation, precipitation scavenging, precipitation, or wave-induced bubble formation leading to the next stage of this iterative cycle).
Additionally, membrane-like phase boundaries are created at each node of the cycle, selectively concentrating organics and metal ions. These metastable boundaries may well have played an essential role in the transition from organic chemistry to biochemistry through the symmetry-breaking segregation of both materials and phases. A potentially critical example of this is discussed in Sections 18.12 and 18.13, where an arguably close-to-ideal microenvironment is created that can support early RNA activity. As an added bonus, the nested hierarchy of microenvironments naturally embodies and functionally supports in a real-world way the majority of other specialized environments and reaction possibilities postulated by other workers in the field (Sections 18.13 and 18.16). But first, we examine new experimental evidence that offers strong support for the claimed advantages of this novel approach to prebiotic chemistry.

18.7 EXPERIMENTAL SUPPORT FOR THE BUBBLE-AEROSOL-DROPLET MODEL IN CHEMICAL EVOLUTION

The first experiments using these ideas in a prebiotic context are only now beginning to be conducted, but with strikingly positive results to date. In an unpublished work, Ruiz Bermejo, Menor Salvan, Osuna Esteban, and Veintemillas Verdaguer (2005) have redone the standard Miller-Urey experiment but utilized surface water induced aerosols. Using a CH₄/N₂/H₂ (40:30:30) atmosphere over liquid water, they used an ultrasonic wave focused on the water’s surface to initiate an aerosol population with an initial droplet size of ~3 μm. The energy source was a standard spark discharge unit, akin to the initial Miller-Urey experiment. The initial results are satisfyingly in line with the advantages predicted by the bubble-aerosol-droplet model. Relative to the standard gas/liquid bulk environments, the introduction of these water-air microenvironments creates:

1. New classes of biochemically important organics (carboxylic and hydroxyl acids along with heterocyclic-like adenine).
2. Chemical yields several orders of magnitude greater than comparable experiments lacking the bubble-aerosol-droplets.

Among the highlights of this experiment are the following:

1. The creation of adenine with a yield 100 times that of previous work. Additionally, Ruiz Bermejo et al. (2005) emphasize that it is free adenine in a soluble form as opposed to previous Miller-type experiments where the adenine was in the form of an unknown precursor. They interpret that the high yield of adenine, using aerosols, as being due to the local enrichment of earlier stage compounds in the liquid-gas interface. Furthermore, the authors state that “this represents a reduction of importance of bulk-solution interfering reactions,” as is predicted by Lerman’s bubble-aerosol-droplet model.
2. Other unexpected heterocyclics are created. For example, Ruiz Bermejo and colleagues clearly demonstrate that hydantoins [C₃H₄N₂O₂] are a real possibility on the early Earth offering, as they suggest a potentially critical role as precursor for prebiotic peptides.* This could be prebiotically important as they are created through bubble-aerosol processes in a “more efficient pathway than the classical base catalyzed hydration of nitriles (under similar conditions of pH and temperature to those of our experiment).”
3. Polyhydroxylated acids are created solely in the bubble-sol version of the experiments, including several sugar-related compounds (such as tartaric and glyceric acid) that have not been seen in comparable prebiotic synthesis experiments.

* Hydantoins can also be precursors to amino acids, but which would racemize easily. That might still be useful in a prebiotic setting . . . who knows? (Steve Benner, private communication, 2006).
4. A range of potentially critical carboxylic acids are also uniquely created in these experiments (as opposed to the standard bulk reservoir approach). Again, quoting Ruiz Bermejo et al:

The succinic and malic acids are members of the Krebs cycle . . . . These carboxylic acids could take part in a primordial variant of the Krebs cycle. This demonstrates that aerosol chemistry also could have contributed to the production of the raw materials for primordial metabolism.

To conclude, Ruiz Bermejo and colleagues state:

As a general conclusion of the results obtained, we can say that the analysis of organic material obtained in presence of aerosol shows greater amounts and greater diversity of molecules than the material obtained using the same conditions without aerosol. The experimental evidence obtained in this work support the hypothesis made by Lerman, that the aerosol droplet behaves as a microscopic chemical reactor that offers the possibility of the concentration, stabilization and transformation of molecules synthesized by means of the energy supplied by the spark discharge. Therefore, we suggest that aerosols could play a significant role in the origin of molecular diversity, evolution, and the origin of life.

Subsequent experimental work by Ruiz Bermejo and colleagues at the Centro de Astrobiologia (Ruiz Bermejo et al., 2007a, 2007b) nicely confirms and expands upon this early work. The number and diversity of positive results in these early experiments is highly encouraging; leading us to expect an increasing number of experiments further exploring the bubble-aerosol-droplet hypothesis. Indeed, Sections 18.12 and 18.13 discuss more speculative inferences from these results that may have additional significant impact on biogenesis.

18.8 CLIMBING THE LADDER OF CHEMICAL EVOLUTION: AN OUTLINE OF THE APPROACH

We will now deal with the above-stated functional requirements in more detail. Starting with the question of basic existence, we will show that the system had a high probability and one that is semi-closed. We then discuss the resulting microenvironments with respect to the molecular scale issues of concentration, chemical selectivity, energetics, and heterogeneous chemistry. Lastly, we explore, at the system level, the potential role of these microenvironments for enabling the transition from planetary organic chemistry to the existence of biochemistry (i.e., biogenesis).

We will reason by qualitative analogy with contemporary Earth processes. Because these processes take place at the molecular level, and are independent of planetary-scale phenomena or specific details of local chemistry, they are expected to be primary ones for any terrestrial-like planet with water, organics, and heavy metals. Additionally, many of the effects are driven by surface tension and will therefore to a first order be independent of the strength of the planet’s gravitational field.

We will address in some detail the extent of these processes on the contemporary Earth, fully expecting them to be just as primary on other water and carbon-rich terrestrial-like planets. Many of our results will be directly applicable to the Archaean and the origin of life on the Earth. Additionally, by applying this work to an early wet Mars, we introduce the idea that the bubble-aerosol-droplet cycle could have been the necessary supporting infrastructure for the origin of life during the Martian Noachian. Obviously, the specific numerical values of an early Martian phenomenology will be different. Work to quantify better these processes for Mars, Titan, Europa, and Enceladus is in progress.

18.9 THE SEMI-CLOSED NATURE OF THE BUBBLE-AEROSOL-DROPLET CYCLE

Because of its surface energy, a lone bubble in a contemporary terrestrial ocean will tend to dissolve on its own. With rise times for such bubbles being 0.13 cm/s, a creation depth of 1 m is the nominal limit from which 100 μm bubbles not stabilized by adsorbed surfactants can be expected to reach the surface. Naturally, smaller bubbles would not be expected to make it at all.
As noted above a bubble becomes a micro-version of the air-sea interface, stabilizing itself through the adsorption of a vast variety of dissolved materials. Blanchard (1975) suggests that a steady state concentration due to this adsorption sets in after about 20–40 s of rising (or traveling) through the water. Most tellingly, for our purposes, in laboratory experiments with artificially made bubbles (<100 \( \mu \)m in sea water) Johnson and Cooke (1980) found that:

Bubble dissolution *always* resulted in the formation of a particle. To provide a control we tried to produce water in which particles could not be produced but did not succeed regardless of the treatment. . . . As the bubble dissolved, striations appeared on the surface, probably a manifestation of film collapse. These lines are well known in monolayer studies and are the result of folding of the surface film.

Along the way to this dissolution, a separate set of experiments (Johnson and Cooke, 1981) followed the time evolution of this dissolution, and found a stabilized set of microbubbles with lifetimes of about 24 h. Not only are the resulting particles highly enriched in organic molecules, but one has the emergence of a membrane-like (organic-rich) film.

Both the stabilized microbubbles and the resulting organic particles can act either as nucleation sites for new bubble formation, be adsorbed by other bubbles, or aggregate with other such objects. Through several generations of bubbling, much of the particulate matter in a local solution will therefore become adsorbed and aggregated. This is a very efficient process, whereby the majority of dissolved organics are adsorbed and concentrated from the surrounding water.

On the contemporary Earth, these organic rich particles are a principal form of food for marine life from a variety of zooplankton in the upper ocean (Baylor and Sutcliffe, 1963; Wangersky, 1974; Sieburth, 1983; Mitchell et al., 1985) to the largest of whales. But in the prebiotic context, without competition from extant life-forms, bubble-created organic-metal rich particulates (and still separate molecular concentrations) would be free to be serially collected by successive generations of bubbles. While some would sink to the sediment, the majority would be taken to the water’s surface. The salient point for cycle continuity is that these same materials will be preferentially formed into the surface of bubbles.

Thus, upon the bubble’s bursting the organics and accompanying surfactants will be further concentrated and released into the atmosphere as aerosols and film-cap drops. By conservation of momentum, when bubbles burst on the surface, ejecting jet drops and film cap drops into the atmosphere, concentrated matter is also ejected into the ocean. Much of this downward-oriented matter becomes the next generation of bubbles. As previously asserted, once organic matter, metal ions, and even mineral surfaces enter into the bubble phase of the overall cycle, they tend to be continuously recycled and worked upon.

But what of the atmospheric components of the overall bubblesol cycle? Further cycle continuity follows from the fact that the impact of the precipitation object (raindrop or snowflake) on the water surface preferentially forms bubbles, returning us to the bubble phase of the bubble-aerosol-droplet cycle (Blanchard and Woodcock, 1957). Snowflakes seem to be particularly good bubble makers, producing bubbles <100 \( \mu \)m (Blanchard, private communication, 1986; Blanchard, 1989; Blanchard and Woodcock, 1957).

There is another, equally critical, cycle-continuity process to consider. Raindrops larger than 100 \( \mu \)m typically produce bubbles with diameters less than 200 \( \mu \)m, whereas (water) droplets of less than 100 \( \mu \)m tend not to directly produce bubbles. Since most aerosols are <100 \( \mu \)m, dry deposition of aerosols may not make bubbles efficiently. On the other hand, organic materials from dry deposition will still be preferentially trapped in the sea-surface microlayer, and immediately reenter the bubblesol cycle by adsorption onto those bubbles created by wind-induced whitecaps and breaking waves. Such waves will be readily available, for wave breaking represents the major loss of (wind) momentum flux in momentum transfer (to oceanic waves) at the air-sea interface (Melville and Rapp, 1985). On the contemporary Earth the most important bubble makers are whitecaps forming at wind speeds exceeding 3 m/s, and with a time-averaged whitecap bubble coverage of roughly 1% of the oceans’ surface at any time.
In summary, precipitation-induced bubbles are an important link in merging the oceanic and atmospheric subcycles of the entire bubble-aerosol-droplet supercycle, while wind-induced bubbles will tend to collect the rest of the material congregating at a sea surface. Hence once organic matter, inorganic ions, and terrestrial and meteoritic particles enter anywhere into the bubblesol cycle, they tend to be continuously recycled, becoming available for the self-organizing chemical-physical processes discussed in the remainder of this paper.

18.10 ENERGETICS: POTENTIAL MECHANISMS OF BUBBLE-INDUCED CHEMISTRIES

In past modeling of prebiotic chemistry, much attention has been paid to reactions in homogeneous aqueous solutions (Chang et al., 1983). In addition, many studies have concentrated on the catalytic processes on the surfaces of solids, especially clays (Rao et al., 1980). However, in the prebiotic context, very little is known of the reactions that occur at the gas-liquid interface, especially those at the surface of bubbles and droplets. As discussed in Section 18.7, a fluid interface determined by phase boundaries can act as a region where the chemistry may take quite different pathways than in the bulk homogeneous environment. Not only are organics and metals selectively concentrated, but there is the potential for relatively anhydrous and short-lived high temperature non-equilibrium regimes. Taken together, these geophysical/geochemical microenvironments allow for new classes

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**FIGURE 18.5** (See color insert following page XXX.) The unique phase-space of sonochemical energetics. On an early terrestrial-like planet there will be great ranges of acoustic cavitation energies; ranging from ocean waves to near-surface geophysically active regions (submarine volcanoes) to meteoritic impacts on an ocean or sea; and of course accompanying all will be the ubiquitous bubble. (From Suslick, K. S. 1989. The chemical effects of ultrasound. *Sci. Am.* 260:80–86. With permission.)
of reactions to occur under what are likely prebiotic conditions. There are four different mechanisms by which such chemical changes may be specifically facilitated inside bubbles:

1. Concentration of organic molecules at the interface due to dipolar hydrophobic-hydrophilic interaction with the two phases. This leaves the molecules oriented in a nonrandom fashion, reducing the entropy of reaction activation between two adjacent molecules. Such specific interphase-induced orientations may play a role in the selection between competing chemical reactions.

2. The release of a water molecule from a condensation process into the aqueous phase at the interface may involve a higher gain in enthalpy and free energy, compared with a reaction in a homogeneous aqueous medium, because the reactants were less hydrated to begin with.

3. The surface compression or shrinkage that occurs on condensation of two molecules on the inner surface of a bubble involves a gain in free energy, similar to carrying out a reaction under hydrostatic pressure. This might lower the free energy of activation of condensation while increasing that of the reverse hydrolysis process. This effect might, therefore, drive the chemical system in the desirable direction from the perspective of chemical evolution since polymerization is in essence a dehydrating condensation process.

4. The rapid contraction of bubbles is associated with adiabatic compression of the internal gas and, therefore, with local heating of the inner surface. This heat will accelerate any chemical reactions at the surface. Early experiments by Fitzgerald et al. (1956) suggested regimes of non-equilibrium chemistry with shock temperatures up to 1000s K or pressures in the kilobar range, while Anbar (1968) was the first to suggest its applicability to prebiotic chemistry.

Only in the past few years, however, have these high non-equilibrium temperatures been confirmed (Brenner et al., 2002). In fact, acoustic cavitation creates regimes of non-equilibrium energetics quite unlike any other and deserves serious further investigation as to its likely prebiotic role. Suslick et al. (1999) and others have shown that these sonochemical hot spots, existing in otherwise cold regimes, can have temperatures of 5000 K, pressures of 1000 atm, and heating and cooling rates of 10 billion °C/s (Figure 18.5). As Suslick (1994) says, “For a rough comparison, these are, respectively, the temperature of the sun, the pressure at the bottom of the ocean, the lifetime of a lightning strike, and a million times faster cooling than a red-hot iron rod plunged into water. Thus, cavitation serves as a means of concentrating the diffuse energy of sound into a chemically useful form.” Indeed, there are indications that the majority of energy released in energetic bubble burstings goes into chemical reactions (Lohse, 2002; Didenko and Suslick, 2002). Because these are rapidly quenched non-equilibrium processes, conventional high-temperature destruction of organics and complex chemistries need not occur. Indeed, these processes may well expedite the condensation reactions (and other endothermic reactions) of interest in chemical evolution that are facilitated or accelerated by any of the other three mechanisms.

Each of these four processes involves the transduction of mechanical into chemical free energy. The first three processes convert surface free energy, generated from mechanical energy (agitation or turbulence), into chemical free energy. The fourth is a thermal mechanism, not involving surface energy, which may accelerate reactions catalyzed by the air-water interface, solid surfaces, or colloidal particles. The latter mechanism may be important because such catalysts also aggregate at the surface of bubbles. Also important with respect to organic reactions on catalytic mineral surfaces are the surface orientating entropy-reducing effects of the first mechanism. We have, therefore, substantial reasons to expect significant changes in chemical behavior in systems with bubbles.

18.11 THE HETEROGENEOUS CHEMISTRY OF AEROSOLS

A detailed overview of the energetics and heterogeneous chemistry available in this phase of the bubblesol cycle follows. As in the case of bubbles, there are substantial reasons to explore the
special chemical environments associated with these micro-Miller bottles (Figure 18.6). A different chemistry than that of bubble phenomena may be associated with aerosols, making them of additional interest in further explorations of the phase space of combinatorial chemical possibilities.

The mechanisms by which specific chemical changes will occur on the outer surface of droplets and liquid aerosol particles are complex. The only trivial catalytic mechanism is the concentration of solutes as the water evaporates. The shrinkage of a droplet involves an increase in surface free energy that results in the cooling of the droplet. The surface orientation effects described above for the inner surface of bubbles will also exist here, leading to enhanced condensation of appropriately oriented species.

There is also the possibility that the dissipation of the high surface energy of a small droplet on impact with bulk water will produce some other form of free energy than heat. Since these droplets are usually charged, the released charges may induce additional types of redox reactions that may not occur in bubbles. Moreover, in the primordial world there would have been a significant flux of ultraviolet radiation to initiate photochemical processes. The interior of the droplet will typically contain a solid nucleus, able to promote heterogeneous reactions (Table 18.1; Turco et al., 1982), and which may in turn be photochemically induced.

These possibilities are further compounded by the many subcycles of phase transitions that these bodies undergo: on the contemporary Earth the condensation nucleus of an aerosol typically undergoes ~10 hydration-dehydration cycles over a 7-day period (T. Graedel, private communication, 1986).

Neglecting, for the moment, the anoxic environment of the primitive Earth’s atmosphere and the still unknown atmospheric chemistry of an early Mars, extrapolation from contemporary aerosols and their chemistry within the aqueous solutions of these atmospheric bodies offers some unique and unusual conditions for metallo-organic chemistry and catalysis:

1. The pH of raindrops is initialized through its equilibrium with atmospheric CO₂ and subsequently modified through the scavenging of aerosols and gases. Sea-salt aerosols ejected from bubbles are slightly alkaline (pH of 7.8–8.3), which is principally due to NaCl.
2. The metal ions accumulated from bubble bursting or atmospheric scavenging, coupled with their relative inability to diffuse out through any organic skins, will lead to the possibility of soaps, foams, and emulsions.

3. Polyvalent metallic ions can increase the surface tension of such films by up to 50%, thus making surface-spreadable organic substances, which would not do so on a pure H\textsubscript{2}O surface.

4. Homogeneous catalysis (including those involving redox reaction) is likely to occur due to transition metals.

5. Heterogeneous catalysis with the solid core is likely to be important. Metal ions as well as clay and mineral particles will be in the core due to atmospheric scavenging. This is substantiated by recent work on aerosols (Murphy et al., 1998) indicating the incorporation of a large range of meteoritic transition metals coupled to high organic content (10–50%). In fact, of the 46 different elements accompanying the organic molecules, the majority of metals were found to be meteoritic as opposed to anthropogenic.

6. The successive hydration-dehydration cycles lead to the likelihood of condensation reactions, accompanied by the possibility of their encapsulation within phospholipid bilayers (as described in the next section).

18.12 ON AMPHIPHILIC BILAYERS AND THINGS TO GO IN THEM

The production of topologically closed amphiphilic bilayers, surrounding a solute rich in other organics and metal ions in non-equilibrium proportions, may result from the rehydration of previously dehydrated organic films (Deamer and Pashley, 1989). Taking the simplest single hydrocarbon chain amphiphiles as a sample case, this requires relatively stringent chemical conditions:

1. An admixture of amphiphiles $> C_{10}$.
3. Hydration time scales sustained for hours to days.
4. Constant high concentrations of amphiphilic compounds, hence requiring a continual source of fresh amphiphilic material (since the decay constant for anything but the simplest molecules is days to weeks).
This is remarkably close to the aerosol-droplet environments described above:

1. Aerosols derived from oceanic bubbles, and with atmospheric scavenging, would be rich in whatever fatty acids, phospholipids, proteins, and other organic compounds were available. Evidence taken from Jaenicke (2005) supports the extremely high percentage of complex organics of all types that will be adsorbed onto (contemporary) aerosols.
2. Such atmospheric bodies would likely have a pH in the range of 7.8–8.3, thus promoting a variety of metallic-amphiphilic soaps.
3. Contemporary aerosols have a lifetime of about 1 week, and in the process go through perhaps 10 cycles of hydration/dehydration.

Recall that in Section 18.9, we saw the formation of a similarly organic rich membrane/film during the bubble phase of the cycle. Therefore, at every stage of the bubble-aerosol-droplet cycle there is the opportunity of not just making a ladder of increasingly complex organic molecules but of “wrapping them up” as well. Following the lead of Deamer and Pashley (1989), subsequent work supports the notion that amphiphilic compounds of meteoritic origin and/or of terrestrial “modification” (in a prebiotic sense) are capable of creating encapsulated microenvironments in which a variety of protometabolic processes and pathways can occur. This is principally due to the work of research groups led by Dave Deamer (Monnard et al., 2002; Dworkin et al., 2001; Apel et al., 2002), Jack Szostak (Szostak et al., 2001; Hanczyc et al., 2003; Hanczyc and Szostak, 2004; Chen and Szostak, 2004; Chen et al., 2004), and Luigi Luisi (Luisi et al., 1993, 1994; Bachmann et al., 1992).

18.13 FROM CHEMICAL EVOLUTION TO BIOGENESIS

Everything above leads directly to the following hypothesis. In spite of considerable effort, no synthesis of RNA has yet been found “in the wild.” Hence, it would seem natural to suggest that RNA synthesis, based on the precursors previously created, might have occurred inside the microenvironments and protocells created by the bubble-aerosol-droplet supercycle. This hypothesis is supported by the fact that these microenvironments naturally produce conditions close to the optimum for RNA activity: a pH of 8–9, high salinity, divalent ions such as Mg, and monovalent such as Na and K (Laura Landweber, private communication, 2000). RNA synthesis involving hydration-dehydration-induced condensation reactions (Zaug and Cech, 1985) could then be supported by these dynamic microenvironments.

Of a more speculative nature, these microenvironments may also support a novel idea on the origins of pre-RNA recently put forward by Simon Nicholas Platts (2004). Platts suggests the existence of an early PAH world,* in which the functional structure of an RNA progenitor was templated by a congruent structure of stacked polyaromatic hydrocarbons (PAHs). These PAHs are themselves hypothesized to have been organized by an interfacial phase separation between the discotic (columnar) aromatic cores of the PAHs and the surrounding water (Figure 18.7). One of the most important consequences of their use as molecular scaffolding is the creation (due to π-π interactions) of a spacing distance of 0.34 nm rise per base pair, the same as RNAs.

The underlying concepts of PAH world have not yet been tested experimentally. But they fit in nicely with the ideas we have elaborated here on the critical role of interfaces as the locales for self-organization (Section 18.1), in particular, the role of the water interface in functionally supporting and geometrically organizing prebiotic chemical evolution. With respect to the prebiotic Earth, recall from Figure 18.4b that PAHs are massively concentrated in just these same air-water interfacial microenvironments.

* Dubbed “PAH world” by Robert Hazen, an excellent historical overview of the development of this model can be found in his new book, *Genesis: The Scientific Quest for Life’s Origins* (Hazen, 2005).
FIGURE 18.7 The PAH–world hypothesis of Nick Platts. (a) Nick Platts’ PAH world hypothesis utilizes the organizing ability of a water interface to facilitate the self-organization of polycyclic aromatic molecules (PAH) into stacks. (b) Once in a stacked and discotic array, the PAHs attract small flat molecules (notably the bases of DNA and RNA) to the edges. Three prebiotic nucleobases are shown hydrogen bonded to hydroxy functions in the edge structures of derivatized neighboring PAH molecules. (c) A molecular backbone forms linking the bases into a long chain. (d) The RNA-like chain of bases separates from the PAHs and folds into a molecule that carries information. (e) Complex assemblages of these chains have the potential to catalyze reactions. (From Platts, N. 2006. Contributions to chemical questions in origins of life. PhD thesis, Department of Chemistry and Chemical Biology, Rensselaer Polytechnic Institute, Troy, NY. With permission.)
Another possibility that these microenvironments might help explain are those hints of a hydrophobic/hydrophilic codon-anticodon correlation (Lacey and Mullins, 1983). In Lerman (1992), we both address these questions in a broader context and describe a class of early experiments, designed by Anastasia Kanavaroti, to test the possibilities of nucleotide polymerization. Given such a situation, it should be easier to make (and make use of) nucleic acids in the more protected confines of a metabolizing, ATP-utilizing, bilayered protocell. For example, the polymerization of ATP cannot occur in water due to electrostatic hindrances, but can do so on an anhydrous surface.

As we have seen, the bubble-aerosol-droplet cycle provides a sufficiently broad range of mechanisms to support in a real-world context, not just the conventional hypothesis of the creation of a RNA world but also the more recent work by the Deamer, Szostak, and Luisi groups (op cit.). Even more generally, the underlying infrastructure we propose might even support the two-origin theory of life put forward by Dyson (1999) and Shapiro (1999): metabolism first, replication second. Indeed, Ruiz Bermejo et al. (2005) based on their initial experiments in support of the bubblesol hypothesis, speculate as to the implications for an early metabolism scenario:

Metabolic type reactions could have had a central role in the processes that gave rise to the origin of life. Currently, the issue about if ‘metabolic life’ could have truly existed and preceded life is controversial. Our experiment with aerosols support the [possibility of the] ‘metabolic life’ hypothesis and demonstrates that the materials for the development of a proto-metabolic system can be synthesized simultaneously with the structural and information system materials. These materials could establish a cycle of reactions as a precursor to intermediary metabolism. (Ruiz Bermejo et al., 2005)

Crossing the great divide from chemical evolution to biological, we can further speculate about the potential relationship between bubbles, bacteria, and Lynn Margulis’ theory of the endosymbiotic origins of eukaryotes (Margulis, 1981). Lerman and Teng (2004a) note that the extraordinary efficiency by which bacteria are collected and concentrated by bubbles (Blanchard and Syzdek, 1970a, 1970b) invites the question: could bubblesol phenomena have helped catalyze the endosymbiotic relationship between prokaryotes that resulted in the development of eukaryotes?

In particular, did the hydrophobic/hydrophilic properties of bubbles concentrate the otherwise disparate bacteria leading to unusual populations at the sea surface, in bubble-created marine snow, and in the sea-salt aerosol?

Analogous to the contemporary world, an Archaean version of marine snow would have provided a superior microenvironment for collections of bacteria to survive, and disparate species of bacteria collected onto a jet drop would undergo a variety of environmental assaults ranging from enhanced UV radiation, to repetitive hydration-dehydration and freezing cycles. Early cell membranes could hardly have remained completely intact during all of this, quite possibly leading to a mixing of bacterial components. Mutations as well would more likely have been induced, to the occasional advantage of “interesting” combinations of cellular components.

18.14 DOES THE ELEMENTAL CHEMISTRY OF LIFE MIRROR THE SEA-SURFACE MICROLAYER?

Due to bubble mass transfer from below, and deposition from the atmosphere (both dry and wet), the concentration of selected organics, metals, ions, and particulate matter can be $10^4$ (or more) greater than the concentration of these materials in the bulk water. Upon inertial bubble formation, it is this surface layer that becomes the inner surface of the bubble. This leads to an intriguing smoking “bubble” of a phenomenological link, correlating sea-surface microlayer concentrations with the chemical composition of organisms across the breadth of life’s kingdoms.

Banin and Navrot (1975) point out that a plot of elemental enrichment factors (the ratio of concentration of an element in an organism to its concentration in the Earth’s crust) versus ionic potentials for four major groups of organisms (bacteria, fungi, plants, and land animals) are fascinatingly
congruent to that of seawater (Figure 18.8). What is most intriguing is the fact that for ionic potentials greater than 10 (sulfur, carbon, nitrogen, and other of the primary biochemical elements), organisms have enrichments of 10–10,000 over the nominal ocean value. This is also the primary range of enrichments due to bubble processes at the sea surface. Hence, there is the strong hint that the enhanced elemental enrichments of living systems are correlated with the sea surface micro-layer enrichments resulting from these bubble processes.

18.15 DO MARTIAN BLUEBERRIES HAVE PITS? CONSEQUENCES AND ARTIFACTS OF “ORGANIC” WEATHER CYCLES ON AN EARLY MARS

We now turn to the more general question of chemical evolution on another terrestrial-like body, and ask whether this cycle could have supported prebiotic chemical self-organization during the Noachian and Hesperian periods on Mars. A warmer early Mars, one with intermittent water but lacking tectonics, offers more limited geophysical/chemical opportunities to support chemical evolution of the type we believe occurred on the Archaean Earth. But for the same chemical physical reasons as on the early Earth, the bubble-aerosol-droplet cycle would likely have existed on an earlier warmer Mars with at least intermittent freestanding bodies of water (Lerman, 2002a, 2004a, 2004b, 2005a, 2006; Lerman and Teng, 2004b).

That the bubble-aerosol-droplet cycle is the predominant one on the contemporary Earth for the concentration and transport of organics, dependent for its initiation only on the Rayleigh-Taylor instability, is a confidence-inspiring measure of its robustness. The fundamental mechanisms of the bubblesol supercycle require only a liquid water-gas interface, and are therefore independent in basic form of the assumptions of specific atmospheric, oceanic, or geological chemistry. Hence, the model is applicable to any planetary body that has, or had, a liquid water-gas interface. And there is increasing reason to believe that the free running water and the hydrology cycles underlying the bubble-aerosol-droplet cycle may have existed on an earlier warmer and wetter Mars (Figure 18.9). This was becoming evident before the era of the Mars Exploration Rovers (Kargel, 2004a, 2004b; Second Conference on Early Mars, 2004). More recently the extraordinary efforts of the MERs Opportunity and Spirit, and their human teams, have provided dramatic evidence for water rich environments in Mars’ geological past (Squyres et al., 2004, 2005, 2008, 2009).

FIGURE 18.9  (See color insert following page XXX.) Possible hydrological cycles on an early wet Mars. Compare to the bubble-aerosol-droplet supercycle outlined in Figures 18.1 and 18.3. (Drawing by Jessica Rhodes after Kargel and Strom, 1996.)
Because bubble processes take place near the surface of water, it matters little if the early Martian oceans were kilometers deep or merely meters. Of course the vapor pressure of water in the Noachian atmosphere would have played a large role in the sizes and timescales of early Martian atmospheric phenomena. And a difference in gravity and atmospheric pressure can make a substantial difference on dynamic processes in an early Martian atmosphere. Not only will the terminal velocity of falling precipitation objects be different, but recent work by Xu et al. (2005) shows that for splashing droplets the surrounding air pressure can change the instability of the edge effects (i.e., the existence of a surrounding crown). But this is more important for droplets falling on a hard surface, and seems less likely to qualitatively affect the substance and nature of droplet-induced bubble formation.

More generally, however, on the tectonically simple early Mars (and one having liquid water only intermittently on its surface), such a complex hydrology cycle may have been the only initiator and supporter of the rapid cycles of concentration, hydration, and dehydration necessary for organic polymerization in the “bulk” quantities necessary for chemical evolution to occur (Lerman, 2002c, 2004b, 2005b; Lerman et al., 2004).

In proposing the functional cycle presented above, we are consistent with the game of minimalist prebiotics, making the simplest strong-principle–based assumptions on the early planets. As an example, one can assume the existence of bubbles on the early Earth and Mars with greater confidence than (say) the widespread availability of a particular montmorillonite clay. And because the existence of these bubbles, cavities, and droplets are such common phenomena in nature, their relevance to chemical evolution is potentially critical, whether on the Archaean Earth, the Noachian Mars, or any other terrestrial-like planetary body.

**FIGURE 18.10** Martian blueberries (NASA. With permission).
One of the more experimentally relevant consequences is the possibility that the Martian “blueberries” (Figure 18.10) discovered by Spirit and Opportunity (Squyres et al., 2004) are nucleated around organic matter or otherwise mediated by organic rich fluids. These concentrated organics may in turn be the result of Martian analogs to the above-described “organic weather” cycles that follow the bubble-aerosol-droplet supercycle.

From their initial discovery, Martian “blueberries” were linked to terrestrial concretions as their most likely analog; with the strong implication that they were similarly a result of Martian sedimentary processes. If Martian “blueberries” are concretion-like objects, then their ubiquity suggests highly efficient formation processes. On Earth, the most efficient of such processes (for ooids to concretions larger than the size of the Martian blueberries) involve organic nucleation sites or organic coatings of mineral cores accompanied by intermittently agitated water. On Earth many of these organic nucleation sites are of biogenic origin. But organic is all that is actually “necessary.” Lerman suggested that this mechanism was also likely for Mars, due to organic complexes likely to have been made by the Noachian equivalent of the bubble-aerosol-droplet cycle (Lerman, 2004a, 2005a, 2006; Lerman et al., 2004; see also Moomaw, 2004).

The potentially crucial role of an abiotic organic component, as opposed to a biological, is highlighted when looking at the terrestrial (Utah) hematite concretions considered closest to the Martian blueberries (Chan et al., 2004, 2005, 2007). A close examination shows that (although a biological nucleation component would not be unexpected) a hydrocarbon-rich fluid component was the likely mediator for their concretions’ formation due to groundwater flow through a permeable host rock coupled to a chemical reaction front.

As on the early Earth, basic organic molecules are expected to have existed on the surface of an early Mars due to deposition by meteorites, comets, and asteroids. But to play such a concretion-forming role requires larger-scale organic “clumps” for nucleation purposes or a high fluid density of longer chain amphiphiles for surface mediation of sedimentation. Either possibility requires a chemical evolution of these primordial organics along with the concentration and aggregation of the resulting higher-order organic molecules. As this paper has clearly shown, all of these evolutionary and aggregating processes are first-order consequences of first-order chemical physics.

The time scales for these processes are fast, being quite compatible with even short timescale intermittent bodies of surface water on an early Mars. Bubble processes occur in seconds to hours; and as shown in Table 18.2 terrestrial timescales for the atmospheric portions of the cycle range from minutes to days (data from Gill et al., 1983). If applicable to an early Mars, this would allow even ultrashort periods of turbulent surface water in a warmer wetter Noachian to support many different cycles of concentration, stabilization, and reactions.

It is thus difficult to imagine an earlier, warmer, and wetter Mars where these processes of organic self-organization did not occur. It is also difficult to conceive of likely alternatives to the bubble-aerosol-droplet cycle as a primary organizer of organic compounds on an earlier Mars.

Another natural consequence of the terrestrial bubble-aerosol-droplet supercycle, and its ability to aggregate organics and metals, are objects that when dried appear fascinatingly akin to the

### TABLE 18.2

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<tr>
<th>Particles</th>
<th>Atmospheric Lifetime</th>
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<td>Aitken particles</td>
<td>1–4 h</td>
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<tr>
<td>Aerosols</td>
<td>4–7 days</td>
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<td>Fog droplets</td>
<td>3 h</td>
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<tr>
<td>Cloud droplets</td>
<td>7 h</td>
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<tr>
<td>Raindrops</td>
<td>3–15 min</td>
</tr>
<tr>
<td>Snowflakes</td>
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</table>

One of the more experimentally relevant consequences is the possibility that the Martian “blueberries” (Figure 18.10) discovered by Spirit and Opportunity (Squyres et al., 2004) are nucleated around organic matter or otherwise mediated by organic rich fluids. These concentrated organics may in turn be the result of Martian analogs to the above-described “organic weather” cycles that follow the bubble-aerosol-droplet supercycle.

From their initial discovery, Martian “blueberries” were linked to terrestrial concretions as their most likely analog; with the strong implication that they were similarly a result of Martian sedimentary processes. If Martian “blueberries” are concretion-like objects, then their ubiquity suggests highly efficient formation processes. On Earth, the most efficient of such processes (for ooids to concretions larger than the size of the Martian blueberries) involve organic nucleation sites or organic coatings of mineral cores accompanied by intermittently agitated water. On Earth many of these organic nucleation sites are of biogenic origin. But organic is all that is actually “necessary.” Lerman suggested that this mechanism was also likely for Mars, due to organic complexes likely to have been made by the Noachian equivalent of the bubble-aerosol-droplet cycle (Lerman, 2004a, 2005a, 2006; Lerman et al., 2004; see also Moomaw, 2004).

The potentially crucial role of an abiotic organic component, as opposed to a biological, is highlighted when looking at the terrestrial (Utah) hematite concretions considered closest to the Martian blueberries (Chan et al., 2004, 2005, 2007). A close examination shows that (although a biological nucleation component would not be unexpected) a hydrocarbon-rich fluid component was the likely mediator for their concretions’ formation due to groundwater flow through a permeable host rock coupled to a chemical reaction front.

As on the early Earth, basic organic molecules are expected to have existed on the surface of an early Mars due to deposition by meteorites, comets, and asteroids. But to play such a concretion-forming role requires larger-scale organic “clumps” for nucleation purposes or a high fluid density of longer chain amphiphiles for surface mediation of sedimentation. Either possibility requires a chemical evolution of these primordial organics along with the concentration and aggregation of the resulting higher-order organic molecules. As this paper has clearly shown, all of these evolutionary and aggregating processes are first-order consequences of first-order chemical physics.

The time scales for these processes are fast, being quite compatible with even short timescale intermittent bodies of surface water on an early Mars. Bubble processes occur in seconds to hours; and as shown in Table 18.2 terrestrial timescales for the atmospheric portions of the cycle range from minutes to days (data from Gill et al., 1983). If applicable to an early Mars, this would allow even ultrashort periods of turbulent surface water in a warmer wetter Noachian to support many different cycles of concentration, stabilization, and reactions.

It is thus difficult to imagine an earlier, warmer, and wetter Mars where these processes of organic self-organization did not occur. It is also difficult to conceive of likely alternatives to the bubble-aerosol-droplet cycle as a primary organizer of organic compounds on an earlier Mars.

Another natural consequence of the terrestrial bubble-aerosol-droplet supercycle, and its ability to aggregate organics and metals, are objects that when dried appear fascinatingly akin to the
so-named “nanobacteria” of ALH 84001 (Lerman 2002a, 2004a, 2005a). These congruent properties include the basic morphology (spheres and sausages), gross chemistry (suites of organics along with metals), and size distributions (nanometers to microns). Whether of biological or bubblesol origin, these striking similarities are due to the universality of the chemical physics involved in the interactions of charge-polarized organic amphiphiles at an air-water interface, coupled to the ubiquity of such scalable macroscopic physical phenomena as the Rayleigh-Taylor instability. The basic morphology and size distributions come from surface tension effects that dominate structure at the micron level while the gross chemistry comes from the concentration effects of the air-water interface for organics, metal ions, and mineral dust. Hence, when designing and interpreting Mars missions looking for evidence of life in Martian paleosols, or when interpreting Martian meteoritic matter akin to the microscopic structures found in ALH84001, the possibility of such abiotic artifacts of past Martian hydrology cycles must be taken into account (Lerman, 2005a).

Thus, it seems likely that the Martian blueberries (and perhaps even the “nanobacteria” of ALH 84001) offer not just evidence of sedimentary processes in a water-rich environment, but hint at the existence of complex past organic weather cycles capable of creating higher-order organics and their aggregates. These same processes of molecular self-organization, which seem to provide the functional requirements for a planetary-scale chemical evolution, are conceivably capable of supporting the autonomous creation of Martian life. Indeed, if the panspermia ideas of Sleep and Zahnle (1998) are correct, that terrestrial life was seeded from an earlier Martian origin, this is all the more relevant. Further work on Mars and the bubble-aerosol-droplet model for prebiotic chemical evolution is under preparation (Lerman, 2006).

18.16 ROBUSTNESS AND DIVERSITY OF EXPLORATIONS THROUGH CHEMICAL PHASE SPACE

This is a new methodological approach, with a strong phenomenological basis in contemporary geophysics/chemistry underlining its potential importance in the reconstruction of chemical evolution. The hypothesis of the fundamental role played by the bubble-aerosol-droplet cycle offers the potential to overcome a number of current stumbling blocks in the field of prebiotic chemistry. In particular, it addresses the problems of selectivity, concentration, and stabilization of organic products in prebiotic chemistry. The bubble-aerosol-droplet cycle affords the possibility of non-equilibrium heterogeneous chemical processes different from conventional solution chemistry, as well as the possibility of coupling a supply of mechanical free energy to chemical processes and reactant concentration mechanisms. It also allows for the possibility of relatively nonaqueous chemical environments within an aqueous medium. Potential condensation reactions are one of the more intriguing consequences. Cycling of hydration-dehydration conditions is easily produced, and with a high surface area to volume ratio for the reactant substrates. A wide range of initial conditions of chemical phase space are sampled, and just as importantly, once organics enter into the bubblesol cycle, they tend to remain within it. All of the above processes are spatially localized and temporally coincident.

Lerman (1986, 1992, 1994a, 1994b, 1996) suggested applying, as global chemical reactor, this contemporary geophysical/geochemical cycle to prebiotic problems. In so doing, it unites and provides a geophysical/chemical basis for a host of other prebiotic studies (Chang, 1993); providing a real-world dynamic framework for the majority of specific chemical model environments developed and suggested by others.

From a historical perspective, Raven and Johnson (2001, p. 66) offer insight on the general concept of bubbles and prebiotic processes. More specifically, Wangersky (1965), in an elegant and far-reaching paper on seawater chemistry, commented in passing that the scavenging ability of bubbles for organics was superior to the similar role played by clays. And Anbar (1968), among others, suggested that bubbles undergoing sonolysis could fix nitrogen from the atmosphere. Polymerization
is a fundamental concern, and the soluble salt substrate model of Chang and Lahav (1982), using hydration-dehydration cycles to efficiently drive polymerization, is naturally supported in the bubble-aerosol supercycle. Regarding the atmospheric component of the supercycle presented here, Woese (1979) first suggested that atmospheric droplets might be useful in prebiotic synthesis; and following the work of Lerman, this was expanded upon by Oberbeck et al. (1991). More recently, Vaida and her collaborators have begun to explore the potential role of the aerosol phase (Dobson et al., 2000; Donaldson et al., 2001, 2002; Tervahattu et al., 2005; Vaida and Tuck, this volume).

The breadth and potential completeness of this overall approach overwhelms Darwin’s “warm little pond” idea, which has resulted in an overly simplistic idea of chemical evolution occurring somehow, somewhere, sometime in an evaporating tide pool. In contrast, the bubble-aerosol-droplet cycle makes optimal use of all the above, and

- Occurs exclusively at the spatial-temporal scales in which heterogeneous non-equilibrium chemistry is most efficiently supported.
- Is coupled to a rapid and robust exploration of chemical phase space.
- Occurs in a coupled geophysical/chemical supercycle that “had-to-have-been” in widespread existence.

Whether on Earth, Mars, Enceladus, Titan, or Europa, in the search for life and its origins in our solar system the global-scale geochemical reactor that is the bubble-aerosol-droplet cycle must be taken into critical consideration. More abstractly, it all stems from the air-water interface and the symmetry-breaking polar nature of the water molecule. Phenomenologically, it reflects the fundamental role of symmetry-breaking interfaces in the support of self-organizing processes as claimed by the author. For this reason, we further suggest that if water is unavailable elsewhere in the universe as the foundation of chemical evolutionary processes leading to an independent origin of life, then other polar solvents must necessarily assume the role.

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WATER AND LIFE
THE UNIQUE PROPERTIES OF H₂O

Reflecting a rich technical and interdisciplinary exchange of ideas, Water and Life: The Unique Properties of H₂O focuses on the properties of water and its interaction with life. The book develops a variety of approaches that help to illuminate ways in which to address deeper questions with respect to the nature of the universe and our place within it.

Grouped in five broad parts, this collection examines the arguments of Lawrence J. Henderson and other scholars on the “fitness” of water for life as part of the physical and chemical properties of nature considered as a foundational environment within which life has emerged and evolved. Leading authorities delve into a range of themes and questions that span key areas of ongoing debate and uncertainty. They draw from the fields of chemistry, biology, biochemistry, planetary and earth sciences, physics, astronomy, and their subspecialties. Several chapters also deal with humanistic disciplines, such as the history of science and theology, to provide additional perspectives.

Features

- Stimulates new investigations among scientists and scholars concerned with the fundamental question of life in our universe
- Provides an overview of the role of water in biochemistry, physical chemistry, earth sciences, and astronomy
- Compares the properties of water with other liquids
- Explores other media that might offer life-generating and life-supporting environments

Bringing together highly esteemed researchers from multidisciplinary fields, this volume addresses fundamental questions relating to the possible role of water in the origin of life in the cosmos. It supports readers in their own explorations of the origin and meaning of life and the role of water in maintaining life.