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A Hybrid Iron/Green-Rust-Urea Model for Prebiotic Chemistry: A Synthesis of Testable Pathways for Planetary Astrobiology

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Abstract

We propose a quantitative, testable framework for abiogenesis that links submarine alkaline vents, which supply H_2 , ΔpH , and Fe/Fe-S catalysis, to subaerial hot-spring fields that provide wet–dry concentration and UV-driven photoredox chemistry. To bridge dilution between environments, we specify mobile "holding pens" (green-rust/iron flocs, silica mats, pumice rafts, and sea-surface microlayer/foam) that concentrate, shuttle, and release prebiotic cargo into shoreline pools. We present an energy ledger that couples geologic gradients and Fe(III)-citrate photoredox to activation currencies (thioesters, condensed P), enabling short-oligomer copying under citrate-buffered Mg^{2*} and protocell membranes compatible with wet–dry cycles. The model makes falsifiable predictions across geology, chemistry, and missions: laminated silica–iron–phosphorus–organic microfacies in Archean analogs; specific rover-detectable co-occurrences on Mars; and plume-analyte patterns on ocean worlds. We provide transport budgets achieving $\geq 10^3$ concentration gain per day, ΔG signposts for keystone steps, autonomous-reactor pass/fail metrics, and an early evolution error-threshold box that motivates short, selectable motifs. The framework paints the big picture while codifying zoomable, lab- and mission-ready tests that can confirm or falsify each link.

Keywords: abiogenesis; greenstone belts; green rust; Fe–S minerals; urea eutectics; rTCA; photoredox; schreibersite; cyanosulfidic chemistry; protocells; astrobiology

1. Introduction

Submarine alkaline vents offer long-lived redox and pH gradients, whereas subaerial hot-spring fields provide concentration, UV-driven photochemistry, and wet-dry ratcheting. Here we synthesize these venues into a hybrid framework anchored in the Dresser Formation (Pilbara Craton), arguing that iron-bearing minerals—green rust (fougerite) and Fe-S phases—formed an inorganic redox architecture biasing carbon flux through non-enzymatic rTCA-like steps.

Figure 1 schematically depicts the coupled venues, transport mechanisms (including a sea-surface microlayer/foam conveyor), and the aerosol pathway; Figure 2 summarizes the stepwise chemical logic separating vent inputs from hot-spring activation and selection.

Figure 1. Hybrid geological setting with SML/foam and aerosol pathway (color schematic) Fig. 1. Vent-derived H₂ and Fe-minerals couple to hot-spring UV/wet-dry chemistry. New elements: a sea-surface microlayer (SML)/foam line at the shoreline and an aerosol pathway that redeposits organic-rich material inland into hot-spring terraces.

2. Theoretical Framework and Mechanisms

2.1 Energy and architecture (vents)

Serpentinization generates H₂ and alkaline fluids, establishing steep pH and redox gradients across mineral membranes. Green rust and Fe-S phases form porous lamellae that shuttle electrons and protons via proton-coupled electron transfer (PCET), acting as inorganic "wires" and semi-permeable barriers.

2.2 Carbon and nitrogen entry

Fe/Ni surfaces reduce CO₂ to formate/acetate under geochemical conditions, seeding C1/C2 organics. Impact-rich episodes and atmospheric processes supply HCN/nitriles and urea; meteorites deliver schreibersite whose corrosion yields

phosphite/pyrophosphate/phosphate—reactive phosphorus species that can accumulate in hot-spring concentrates.

2.3 Anabolic routing and redox reset

Transition metals promote non-enzymatic rTCA-like interconversions among succinate, fumarate, malate, oxaloacetate, and citrate. Once citrate appears, Fe(III)-citrate photochemistry provides a light-driven Fe(III)/Fe(II) cycle that periodically restores reducing equivalents under early-Earth UV/visible bands. Primer citrate can arise non-photochemically at trace levels via metal-promoted rTCA sequences on mineral surfaces—notably Fe/Zn/Cr-assisted steps shown to generate multiple Krebs intermediates [Muchowska et al., 2017; Muchowska et al., 2019]; wet-dry cycling then concentrates these traces to kickstart the photoredox-reset loop.

2.4 Activation chemistry in urea eutectics

Urea-rich eutectics and cyanate routes enhance phosphorylation, mobilize phosphorus from minerals, and assist condensation (e.g., peptide linkages). Wet-dry cycles in silica-rimmed pools ratchet complexity, concentrate reactants, and sift products by phase behavior. Plausible sources include meteoritic/exogenous input and interstellar ice reservoirs (e.g., Murchison; Perrero, 2023), impact-hydrothermal synthesis on Fe-clays [Pastorek et al., 2019], atmospheric/microdroplet pathways (CO₂+NH₃ at air-water interfaces; Song et al., 2023), and cyanamide hydrolysis in UV-lit ponds; urea eutectics also stabilize and activate phosphorylations.

2.5 Transport and the "water problem": proposed mechanism

We propose a vent-to-shore logistics chain: (i) Fe-oxyhydroxide/green-rust flocs and silica gels near vents scavenge organics and reactive phosphorus; (ii) floc aggregates and pumice/ash rafts act as buoyant carriers; (iii) tidally modulated shelf currents and storm waves advect these carriers toward emergent greenstone coasts; (iv) in the surf/swash zone, sorbed cargo is released into hot-spring catchments and trapped in sinter terraces. This "holding-pen conveyor" mitigates dilution by combining sorption, protection, and episodic

2.6 Seafoam/SML transport link (addition to Section 2.5)

Bubble scavenging and foam lines at emergent shorelines act as dynamic concentrators and shuttles. As bubbles burst, organic-rich aerosols carry SML cargo inland and into hot-spring terraces, while persistent foam lines trap and slowly leak sorbed material into pool networks.

Figure 2. Process flow with inputs separated (color)

Fig. 2. Vents (blue) supply gradients and C1/C2 feedstock; hot springs (amber) supply UV activation and wet–dry selection. Shared nodes (gray) operate in either setting or at the interface. Bottom band: transport via iron-rich flocs, silica mats, pumice rafts; foam/SML & aerosols shuttle cargo shoreward.

2.7 Proto-RNA options in the hybrid setting.

This box compares candidate information polymers—RNA and plausible predecessors (TNA, GNA, PNA)—against the geochemical realities of the hybrid vents↔hot-spring framework. We emphasize monomer plausibility, backbone stability under UV/wet-dry cycles, and compatibility with Fe-bearing minerals, urea eutectics, and citrate buffering.

- RNA (ribose-phosphate backbone): Pros: mature literature on nonenzymatic copying; phosphate chemistry meshes with reactive-P pool; Mg²+-mediated catalysis can be moderated by citrate; bases accessible from cyanosulfidic chemistry. Constraints: ribose stability and selective ribose formation; strand separation and Mg²+-induced membrane stress must be managed.
- TNA (threose-nucleic acid): Pros: simpler sugar (C4) may be easier to access; forms stable base-paired duplexes; potential better performance in high-salt/variable pH.
 Constraints: pathway integration with phosphate activation and transition to RNA must be explained.
- **GNA** (glycol nucleic acid): Pros: very simple backbone (C2); robust duplexing; potential tolerance of salts and temperature swings. Constraints: base-pairing rules and stepping stone to RNA require clear interconversion chemistry.
- PNA (peptide nucleic acid): Pros: peptide-like backbone stable to hydrolysis; synergy
 with early short peptides. Constraints: prebiotic monomer availability and coupling
 chemistry under wet-dry cycles are less established; membrane permeability of
 monomers may be limiting.

Working hypothesis: a TNA→RNA transition is plausible—TNA (or GNA) supports initial template copying and selection under harsher cycles; once citrate buffering, Fe(III)—citrate photoredox, and reactive-P management are established, RNA becomes competitive and eventually dominant.

2.8 Nonenzymatic copying under hybrid cycles

We propose a day-night copying protocol synchronized to the hybrid venue. Short activated (proto)NTPs (e.g., imidazole/2-aminoimidazole activated) accumulate in urea-rich eutectics and concentrate in wet-dry phases on silica-rimmed terraces. Copying proceeds on mineral/lamellar surfaces (green rust, Fe-S, clays) or inside fatty-acid vesicles with citrate-buffered Mg²+ to enable catalysis while protecting membranes.

Day (UV/light): Fe(III)—citrate photoredox regenerates reducing equivalents; moderate temperatures favor primer extension on templates; partial strand destabilization by ionic shifts/photothermal heating aids transient dehybridization at short stretches.

Evening-Night (cooling/desiccation): Wet-dry concentration drives ligation and capping; partial desiccation promotes activation chemistry (cyanate/urea routes).

Rehydration (pre-dawn): Fresh inflow re-primes cycles; vesicles grow and divide at film boundaries, passing compositions and emerging oligomers forward.

Target performance: >50% copying fidelity over 10–20 nt fragments under citrate-buffered Mg²⁺, with compatible vesicle integrity.

Design controls include: (i) salt/urea windows that keep vesicles intact, (ii) mineral vs. in-vesicle copying comparisons, and (iii) tests of strand displacement or short-segment 'nicking' to achieve separation without enzyme-level melts.

Table 1. Energy ledger and coupling in the hybrid framework

Step / Module	Energy Currency	Source / Driver	Directional Cue	Feasibility Notes
Serpentinizatio n gradients	ΔpH, H₂ (redox)	Vent fluids; mineral membranes	Proton/electro n gradients	Long-lived geologic battery; feeds ET/PCET across Fe/Fe-S lamellae

CO₂ → C1/C2 fixation	Reducing power (H₂/Fe²+)	Fe/Ni surfaces at vents	Surface catalysis	Formate/acetat e yields seed carbon entry; pairs with Fe mineral ET
rTCA-like interconversio ns	Redox equivalents	Fe/Mn/Ni catalysis	Sequential metal-mediate d steps	Succinate↔ma late↔oxaloace tate↔citrate routing without enzymes
Photoredox reset	Fe(III)/Fe(II) cycling	Fe(III)-citrate + UV/visible	Light–dark rhythm	Restores reducing power daily; tuned to early-Earth UV window
Activation chemistry	Thioesters; condensed-P (polyP)	Urea eutectics; reactive-P (schreibersite)	Wet-dry concentration	Phosphorylatio n/condensatio n for ligation, peptide bonds
Copying & ligation	Activated (proto)NTPs	Imidazole/2-a minoimidazole; cyanate/urea	Template + film/vessel crowding	Short-segmen t copying with citrate-buffere d Mg ²⁺
Compartment dynamics	Mechanical/ch emical work	Osmotic growth; film shearing	Growth-divisio n cycles	Encapsulation, inheritance of catalytic cargo (compositional heredity)

Box 1. Error threshold and early selection windows

Replication accuracy sets an upper bound on selectable genome length. If per-base fidelity is p, the per-genome fidelity is $P = p^L$ for length L. To maintain an informational lineage against a competitor with selective advantage κ (>1), a rough criterion is $P \ge 1/\kappa$. Early systems

therefore favor short templates and motifs; selection acts on many short, partially overlapping segments rather than one long perfect copy.

Interpretation: with p \approx 0.90 and modest $\kappa\approx$ 1.2, L_max \approx 11 nt; with p \approx 0.98 and $\kappa\approx$ 1.2, L_max \approx 56 nt. This supports a trajectory where short oligomers (10–30 nt) become selectable first; amplification and error-correction (templated ligation, sequence-biased adsorption, compartment selection) progressively allow longer motifs.

2.9 Membrane recipe compatible with Mg²⁺/Fe²⁺ and wet-dry cycles

We specify a practical protocell membrane compatible with copying chemistry and the hybrid environment:

- Composition: fatty acid + fatty alcohol mixtures (e.g., oleic acid + oleyl alcohol, 7:3–8:2 molar) to boost stability and reduce leakage.
- **Buffering: citrate-buffered Mg²**+ (1–10 mM free Mg²+ with equimolar citrate) to enable template copying while protecting membranes; Fe²+ kept low (≤0.1–0.5 mM) during in-vesicle copying, higher at mineral interfaces.
- Salinity: tolerate moderate salts (Na⁺, K⁺) with alcohol co-lipids; adjust osmotic gradients to drive **growth/division** during rehydration and film shearing.
- **Permeability:** activated (proto)NTPs permeate slowly; uptake aided by wet-dry cycling and transient defects at film boundaries; retention of short oligomers favored.
- **Division:** film-boundary shearing and rehydration pulses produce **growth-division** without enzymes; compositional heredity maintained when catalysts/cofactors partition with lipids.
- **Membrane-genome coupling:** copying can occur **on** mineral films (green rust/Fe-S/clays) or **in** vesicles. Citrate moderates Mg²⁺ reactivity so both membranes and templates survive; short cationic peptides (if present) can stabilize duplexes and promote strand separation.

3. A day-in-the-life of the hybrid system

Sunrise: UV penetrates shallow hot-spring pools; Fe(III)—citrate complexes photoreduce to Fe(II), resetting the redox state. Urea eutectics concentrate phosphate; cyanate routes activate carboxylates and amines. Late morning: Fe-rich flocs and sorbed organics from the shelf mix with spring waters, releasing cargo into silica-lined basins. Afternoon: rTCA-like interconversions proceed on mineral surfaces; short peptides and Fe-S clusters form and bind to lamellae, improving local electron transfer. Evening: Evaporation concentrates solutes; vesicles nucleate and encapsulate subsets of the network. Night: Partial desiccation favors

condensation; weakly bound products crystallize or precipitate. Pre-dawn: Fresh discharge rehydrates the basin; vesicles grow osmotically, divide at film boundaries, and compositional heredity carries to the next cycle.

4. Falsifiable Predictions, Homochirality, and Astrobiology

4.1 Testable predictions and autonomous criteria

While the schematic pathways highlight clean routes, prebiotic mixtures are chemically crowded; a key experimental challenge—and criterion of plausibility—is demonstrating that the proposed rTCA-like steps, photoredox reset, and copying chemistry operate with acceptable yields without being quenched by competing side reactions.

Co-localization: In Dresser sinters, lamina-scale co-occurrence of silica + Fe phases +

reactive-P residues + organo-mineral films, with Fe/C/N isotope offsets indicative of photoredox and scavenging cycles.

Logistics: Reactive-transport models that—given Archean currents and particle kinetics—achieve ≥10³ concentration gains relative to open-ocean dilution.

Autonomy: Closed-loop, pre-registered reactors that apply only geologically justified forcings yet show net accumulation, regeneration of inputs, and robustness to perturbation.

4.2 Homochirality: mechanism and amplification

We consider magnetochiral anisotropy (MChA) on magnetized magnetite/hematite under UV as a source of a small enantiomeric excess (ee). MChA arises when a magnetic field and circularly polarized light—or magnetization-induced chiral electronic transitions at a surface—break mirror symmetry in excitation pathways, giving rate differences for L vs. D enantiomers. Although the primordial ee is expected to be small, amplification is plausible via enantioselective adsorption on mineral faces, dissolution—crystallization cycles during wet—dry periods, and kinetic resolution within autocatalytic subnetworks.

Box 2. Homochirality: small bias + crystallization cycles Mechanism: a tiny magnetochiral or surface-induced rate bias (≤0.5% per cycle) seeds enantiomeric excess (ee). Crystallization-dissolution cycling (including abrasion/ripening in wet-dry) acts as a non-linear amplifier, preferentially recycling the more soluble enantiomer and enriching the less soluble one. Coupling: foam/SML films and silica terraces provide repeated cycles and gentle grinding/evaporation environments; mineral faces can mediate enantioselective adsorption.

Figure 3. Homochirality amplification (simulation)

Fig. 3. Simulated ee growth over cycles with a small per-cycle bias and crystallization-type amplification. Parameters illustrative; qualitative behavior shows threshold-crossing from ~1% to tens of percent ee.

Figure 4. Conceptual UV action spectrum for Fe(III)—citrate and thin-film effects
Fig. 4. Normalized conceptual curves: relative Fe(III)—citrate photoredox effectiveness vs.
wavelength (solid) and a thin-film enhancement factor (dashed). Exact band maxima depend
on solution chemistry and early-Earth UV; curves are schematic for study design.

4.3 Origin of information: from compositional heredity to templates

Before templated polymers dominate, vesicle populations can inherit functional compositions (lipids, cofactors, short oligomers) that bias metabolic flux. Information-theoretic metrics—mutual information between parent and daughter compositions—provide objective heredity measures. Sequence-dependent binding to Fe/FeS/green rust can then select oligomer motifs that improve electron transfer or catalysis; under the same UV/wet-dry windows, template-assisted ligation crosses fidelity thresholds for short lengths, bridging toward genetic replication.

4.4 Implications for astrobiology (Mars and ocean worlds)

Table 2. Field and	d mission ch	necklist for t	talsificat	tion/con	firmation
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Target environment	Prediction / Signature	Measurement	Instrument examples	Decision rule
Pilbara (Dresser) sinters	Lamina-scale co-localization : silica + Fe phases + reactive-P	Mineralogy, P-valence, organics; Fe/C/N isotopes	Micro-Raman; μ-XRF/ToF-SI MS; NanoSIMS; Fe/P XANES	Presence with isotopic offsets supports H1/H10;

	residues + organo-films			absence in suitable facies disfavors
Pilbara shoreline microfacies	Foam-line textures with Fe-rich phases and organo-films	Textures + organics + Fe/P mapping	Optical petrography; Raman; μ-XRF; FTIR	Co-occurrence consistent with SML/foam H13
Mars silica sinter facies	Silica + Fe phases + reduced-P (phosphite/pyr ophosphate) + organo-films	Elemental maps; organics; P-valence proxies	PIXL, SHERLOC, SuperCam/Ra man, LIBS; sample return	Co-localization strengthens hybrid model; repeated nulls weaken
Mars coastal analogs (if present)	Shoreline deposits with foam-line analog textures	Textures; organics; Fe/P mapping	Mastcam-Z imaging; SHERLOC/PIXL spot checks	Positive suite bolsters SML transport
Europa/Encela dus plumes	Fe-bearing nanoparticles; urea/nitriles; organo-P; C1/C2 patterns	In-flight MS; dust impact MS; isotopes	INMS-class MS; dust analyzers (e.g., SUDA-type)	Joint detection pattern consistent with hybrid feedstock
Modern SML/foam (process analogs)	Enhanced interfacial photoproducts vs. bulk	UV action spectra; product distributions	Benchtop UV-vis; LC-MS; surface tensiometry	Higher yields in films vs. bulk support H13

Table 3. Instrument observables and detection windows (illustrative)

Target / Signature	Instrument	Modality	Spatial scale	Representa tive LOD /	Hypotheses

				Notes	
Reduced P (phosphite/ pyrophosph ate) with Fe phases	PIXL (Mars 2020)	X-ray fluorescenc e mapping	~100 μm pixels	Elemental P, Fe maps; P-Fe co-localizat ion; valence via context (pair with Raman/SHE RLOC)	H1/H10
Aromatic/ali phatic organics bound to Fe-oxides/g reen rust	SHERLOC (Mars 2020)	Deep-UV Raman & fluorescenc e	~100 μm	Organics detection; aromatic bands; context with PIXL	H1/H10/H13
Fe mineralogy & organics (phase ID)	SuperCam / Raman-LIB S	Raman spectrosco py / LIBS	mm-scale	Magnetite/ hematite; LIBS ratios; organic bands (if present)	H1/H10/H13
Silica-Fe-o rganics microfacies in sinter	Raman / µXRF / FTIR (lab or rover)	Vibrational + microprobe	10-100 μm	Co-located silica and Fe-oxide bands; lamina textures	H1/H1O
Urea, nitriles, organo-P, Fe-bearing nanoparticl es	Plume MS (Europa/En celadus)	In-situ mass spectromet ry	Bulk plume / particles	Exact masses & fragments; dust impact MS for nanoparticl es	H2/H4

SML/foam residues analogs; (surface-ac tive organics, Fe) Lab analogs; LC-MS; UV-vis; tensiometry	Lab assays	Film / bulk	Thin-film vs bulk yield; surface activity; distinct compositio n	H13
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Mars: Target silica sinter and hydrothermal facies for lamina-scale co-localization of silica + Fe phases + reactive-P valence states (phosphite/pyrophosphate) and organo-films. Instruments: Raman/LIBS/PIXL/SHERLOC to probe P valence, organics, and Fe mineralogy; prioritize sample return from such facies.

Europa/Enceladus: Plume sampling for Fe-bearing nanoparticles, reduced P species, urea/nitriles, and small organics consistent with CO₂→C1/C2 reduction and cyanosulfidic feeds; isotopic patterns indicative of redox cycling.

Titan: Tholin-rich organics coupled with episodic liquid-water environments (impact melts or cryovolcanism) could provide cyanosulfidic inputs; look for organo-phosphorus species and Fe-bearing catalysts in transient aqueous settings.

4.5 Seafoam/SML predictions (H13)

H13: Seafoam/SML microreactors enhance interfacial photoredox and deliver organics to terrestrial basins.

Predictions: (i) SML-mimic films yield higher interfacial photoproducts than bulk for Fe(III)-carboxylate analogs; (ii) foam-line microfacies preserve co-localized Fe phases with organo-films at lamina scale; (iii) aerosol residues carry SML surface-active signatures distinct from bulk dissolved organics.

5. Methods and Simulation

5.1 Transport and concentration model

We outline a minimal reactive-transport accounting for the "holding-pen conveyor" (iron-rich flocs, silica mats, pumice) plus SML/foam and aerosol shuttles. Let C_0 be the concentration at source; the effective concentration delivered to hot-spring pools after one daily cycle is: $C_0 = C_0 \times F_0 \times F_0$

5.2 ΔG accounting and energy coupling

We provide a back-of-envelope method to assess feasibility and directionality for keystone steps. For each reaction, compute $\Delta G = \Delta G^{\circ}(T) + RT \ln Q$ under the relevant composition and temperature; then indicate the driver that sets Q (light, wet-dry, adsorption). Signposts guide where to spend experimental effort: set light spectra and evaporation schedules to push Q in favorable directions; use mineral/film adsorption to bias activities; and account for coupled removal (encapsulation, precipitation) to make otherwise marginal steps proceed.

5.3 Homochirality amplification

The simulation for enantiomeric excess (ee) growth over cycles uses the recurrence relation: $ee_{n+1} = ee_n + b + \alpha \cdot ee_n \cdot (1 - ee_n^2)$, with illustrative parameters $ee_0 = 0.01$, b = 0.002/cycle, a = 0.08. The curve is conceptual and not a fit to experimental data, but demonstrates the threshold behavior.

5.4 Fe(III)-citrate photoredox spectrum

The conceptual action spectrum is constructed from Gaussian-like bands to represent Fe(III)-citrate photoredox effectiveness and a qualitative thin-film enhancement factor. These curves are schematic for study design and are not measured spectra; exact maxima depend

on solution chemistry and the UV environment.

5.5 Autonomous reactor criteria

The goal is a "hands-off" demonstration in which only geologically justified forcings are applied (light spectrum/intensity, temperature ramps, fluid flows, pH-buffering minerals), with no manual rescues or mid-course reagent additions.

Allowed forcings (pre-registered): Spectrally defined UV/visible illumination; temperature ramps; flow-through of vent-analog and rain/steam-condensate solutions; and mineral cartridges.

Primary metrics (quantified over N cycles): Net mass balance; regeneration of inputs; robustness to perturbation; and compositional inheritance.

Pass/fail thresholds: A run PASSES if ≥2 of the primary metrics are met simultaneously. A run FAILS if any manual rescue is required or if any metric trends monotonically to zero.

5.6 Concise Methods

Autonomy metrics: Implement the metrics noted above by pre-registering experimental conditions and logging all interventions.

Membrane robustness checks: Use fatty acid + alcohol (7:3–8:2) mixtures and verify vesicle integrity across cycles by dye-leak or DLS. Ensure Mg²⁺/Fe²⁺ are within vesicle-friendly windows.

Copying assay windows: Buffer Mg²⁺ with citrate to a free Mg²⁺ of 1–10 mM. Run day–night cycles with a light period for photoredox and primer extension, and an evening/night phase for concentration and activation.

Sensitivity propagation: Draw N=10³-10⁴ samples of the six factors in the transport model to compute the distribution of C_pool/C₀. Report the median and 5th-95th percentiles. Rank-order sensitivity by partial rank correlation or by variance contribution.

6. List of Abbreviations

BIF: Banded Iron FormationDAP: Diamidophosphate

• ET/PCET: Electron Transfer / Proton-Coupled Electron Transfer

• **GA:** Graphical Abstract

- **HCN:** Hydrogen cyanide
- MChA: Magnetochiral anisotropy
- NTP: Nucleoside triphosphate (or activated proto-NTP)
- **rTCA:** reductive Tricarboxylic Acid (cycle)
- **SML:** Sea-Surface Microlayer

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