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Polyelectrolytes with uniform structure are universal for Darwinism

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The polyelectrolyte that supported Earth's first Darwinism was RNA

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A relatively narrow date when life on Earth originated prebiotically

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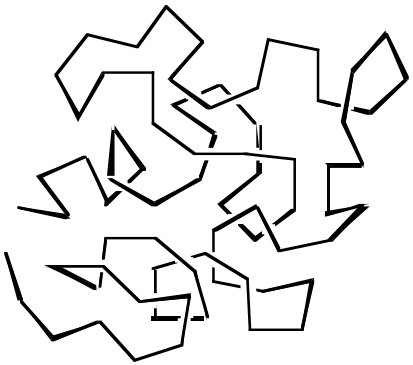
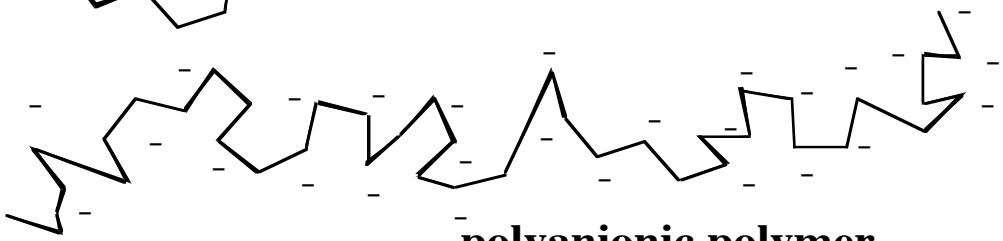
What does a repeating backbone charge do for informational molecule?

1. Keeps the biopolymer dissolved (in water).
2. Backbone-backbone coulombic repulsions force strand-strand contacts to Watson-Crick edges of bases (**= A:T G:C pairing rules**).
3. Polyanion discourages folding, allows templating



neutral polymer
Radius = length $^{(1/2)}$



polyanionic polymer
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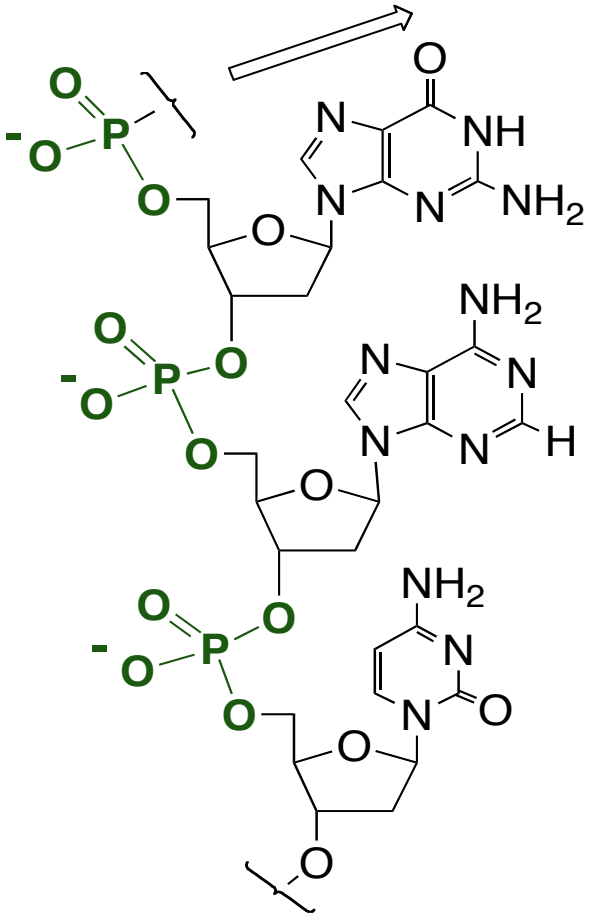
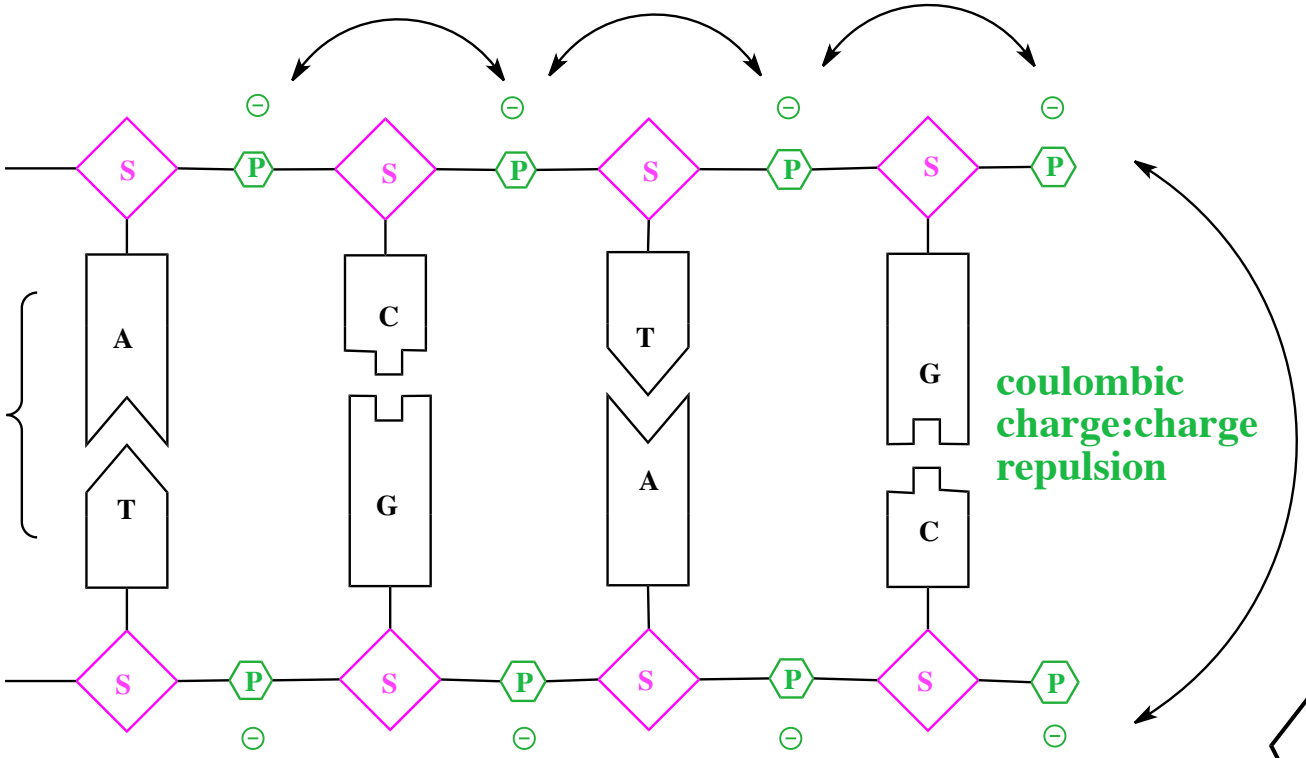
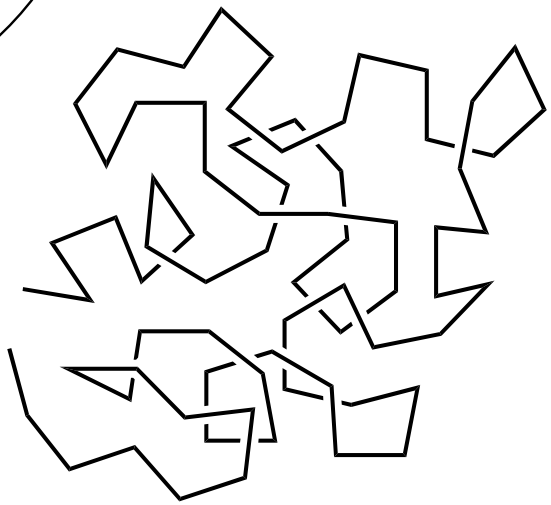


Benner & Hutter, D. (2002) Phosphates, DNA, and the search for nonterran life. *Bioorg. Chem.* **30**, 62-80.

Repeating charge (polyelectrolyte) on backbone discourages folding

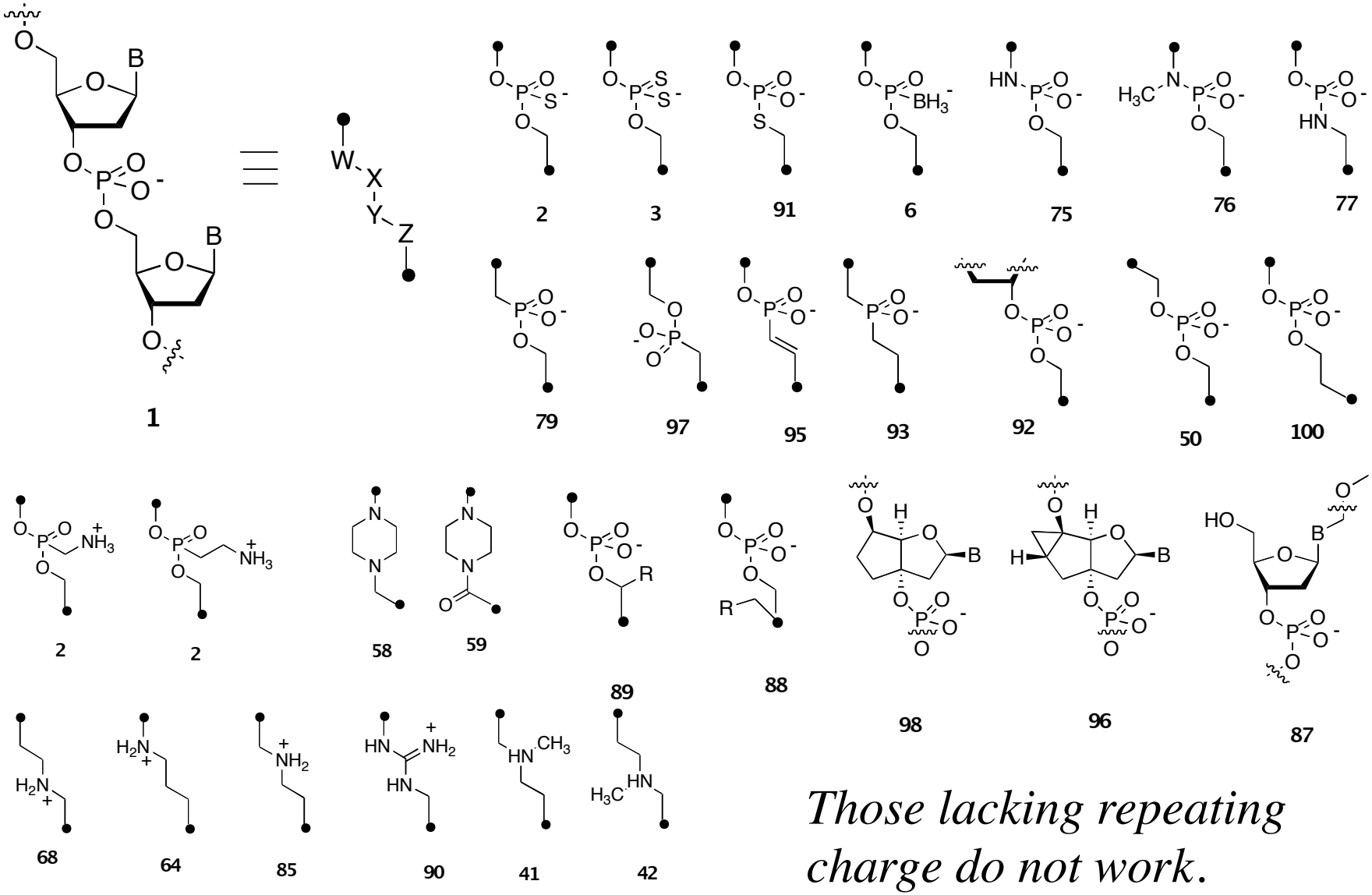


Coulomb's law predicts that the backbones repel. ***Duplex is more stable in high salt.***



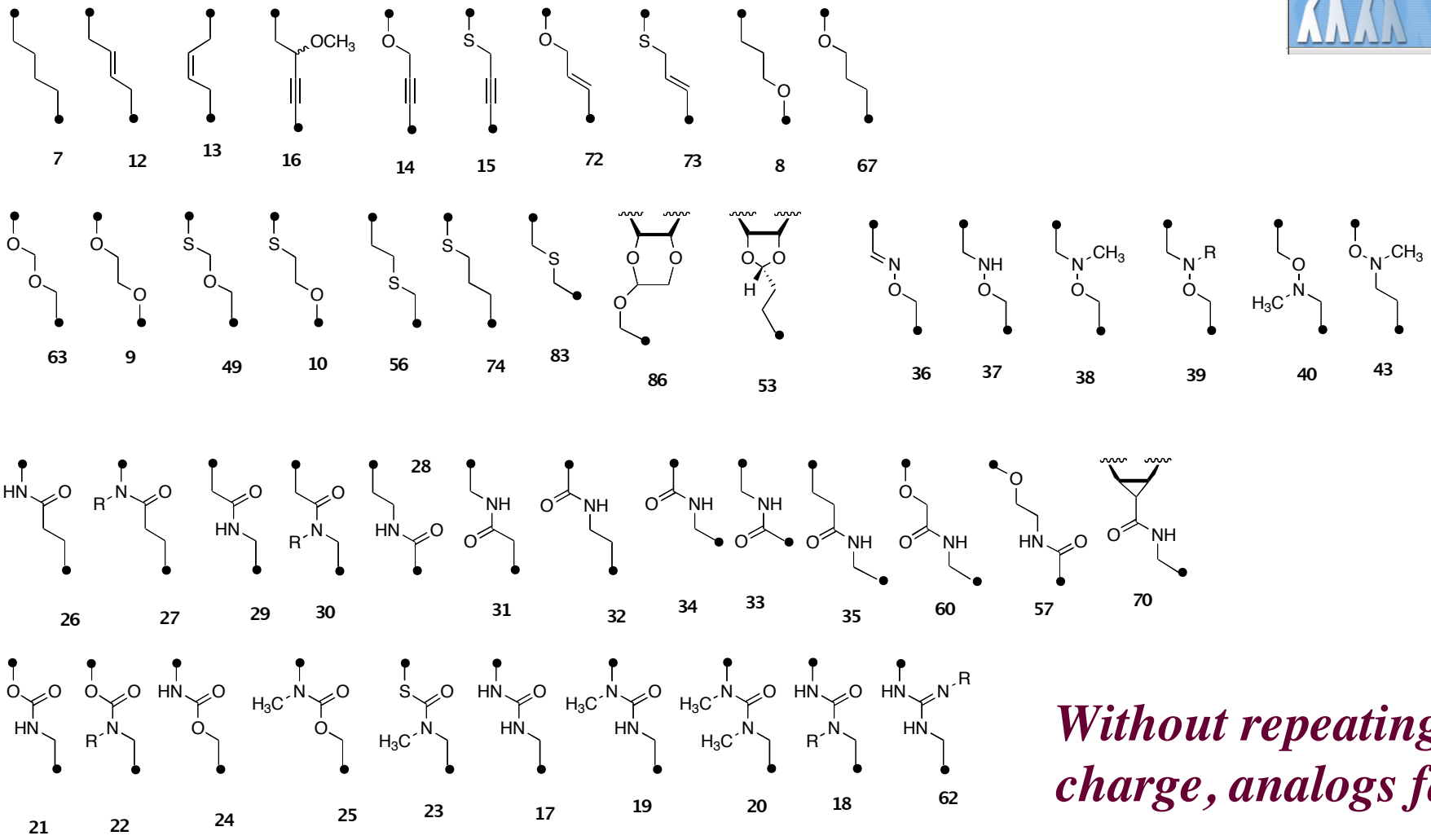
This is needed for templating

Experiments in addition to theory. Synthetic biologists made many DNA variants



Those lacking repeating charge do not work.

Synthetic biologists made many DNA variants



Without repeating charge, analogs fail

Benner & Hutter (2002) Phosphates, DNA, and the search for nonterran life: A second generation model for genetic molecules. *Bioorg. Chem.* **30**, 62-80.

Darwinism needs backbone charge

Must be able to **change** sequence to change **information**…
Without changing its physical properties sufficient to impact its performance in processes involved in inheritance.
Such as: its solubility, its molecular recognition, reactivity
Lessons from chemistry: Such systems are scarce.

Proteins, polysaccharides, abiological polymers most every other class of molecules, physical behavior changes dramatically with structure changes, even small ones

Sickle cell hemoglobin, 1 amino acid change, precipitates

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The repeating backbone charge of DNA so dominates its properties that nucleobase replacement is only a minor perturbation.

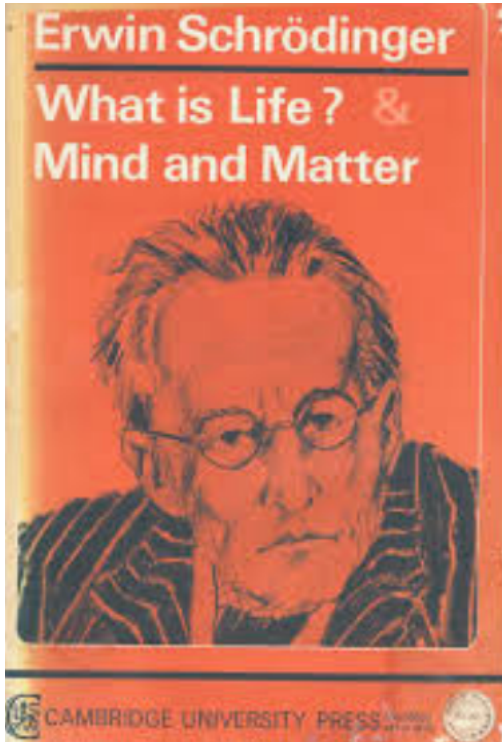
Sickle cell hemoglobin, 1 amino acid change, precipitates

Structural requirements for Darwinism = life

Regardless of origins, in water, Darwinism will be supported by a biopolymer with a repeating backbone charge (negative or positive)
Need to keep physical properties constant upon information change

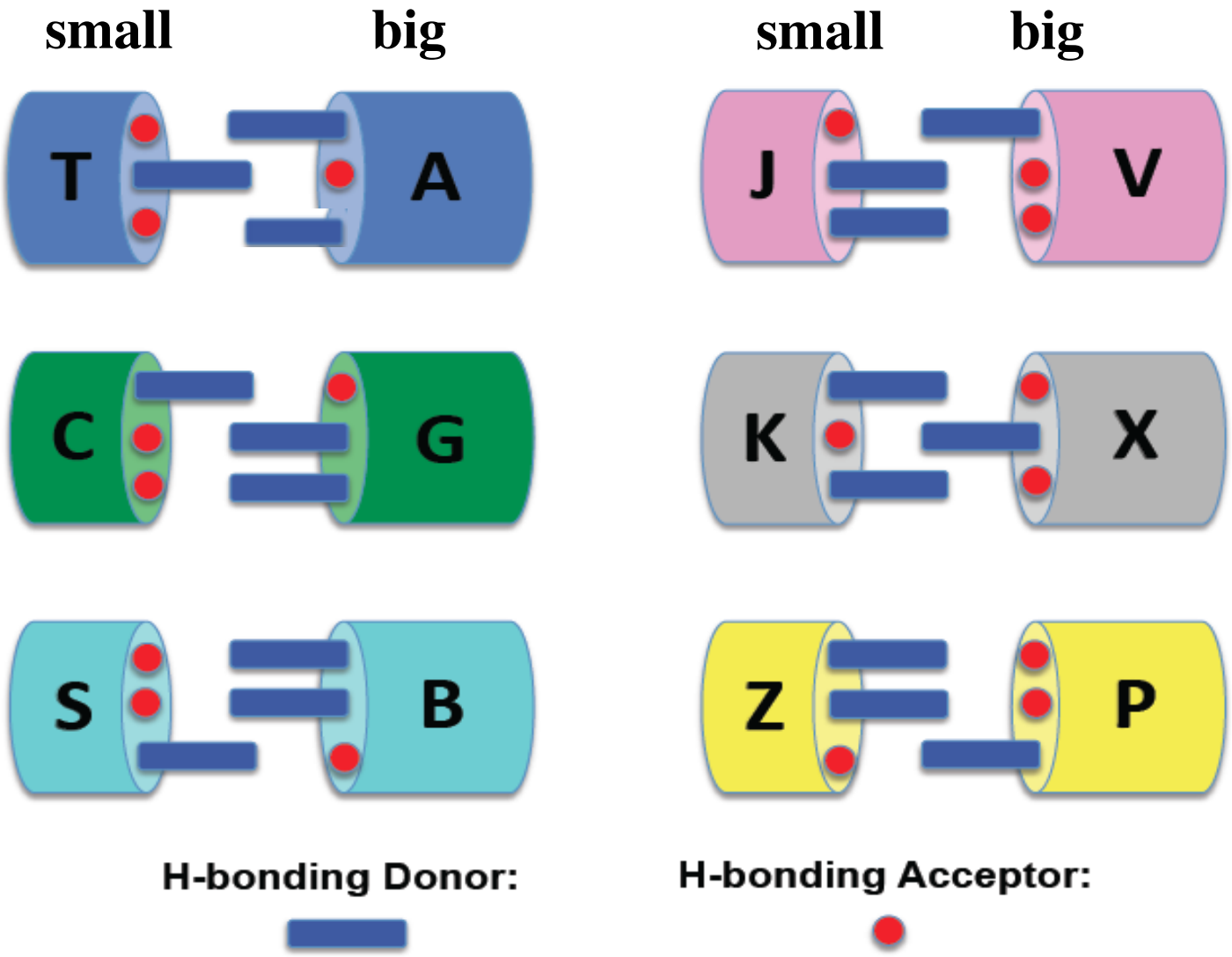
We also must keep *structure constant* upon information change
Schrödinger aperiodic crystal criterion for genetic biopolymers

Schrödinger in 1943 knew nothing about DNA.
But he knew that simple binding cannot guarantee fidelity of information transfer needed for biology. For that, Schrödinger needed the physics of phase transitions. For *that*, exchangeable informational building blocks must all have the same size/shape. They must all fit in an **aperiodic crystal structure**.



Two criteria for an informational biopolymer
(a) Polyelectrolyte backbone
(b) Exchangeable units have same size/shape

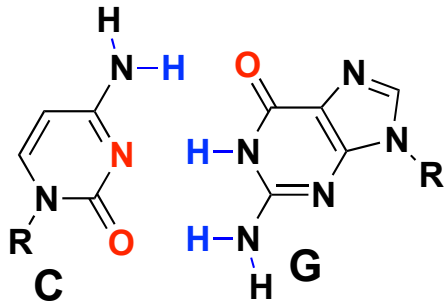
We have synthesized new Darwinian systems following these rules ("alien Darwinism")



Orthogonal binding

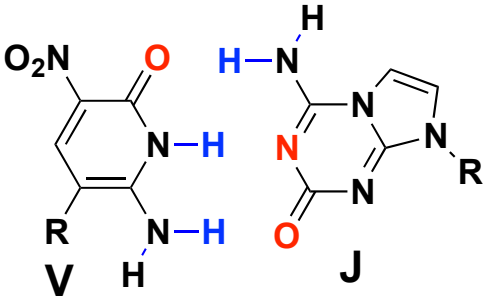
We synthesized new Darwinian systems following these rules ("alien Darwinism")

pyDAA
Donor
Acceptor
Acceptor



puADD
Acceptor
Donor
Donor

pyADD
Acceptor
Donor
Donor



puDAA
Donor
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Acceptor
Donor
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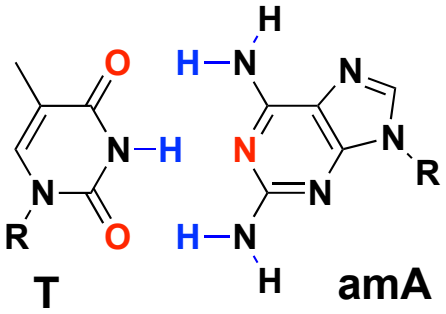

puDAD
Donor
Acceptor
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pyDAD
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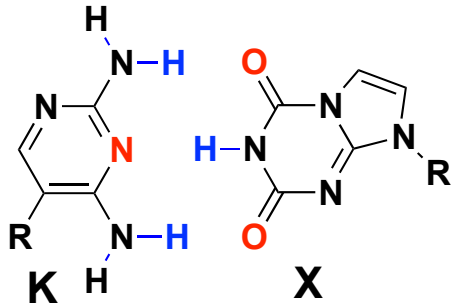
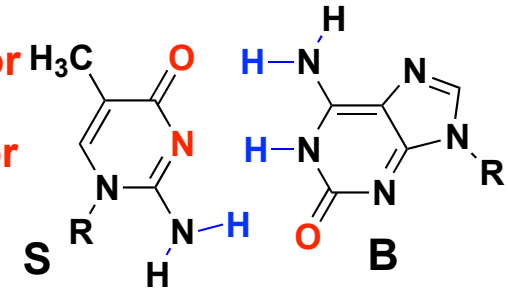
Acceptor
Donor



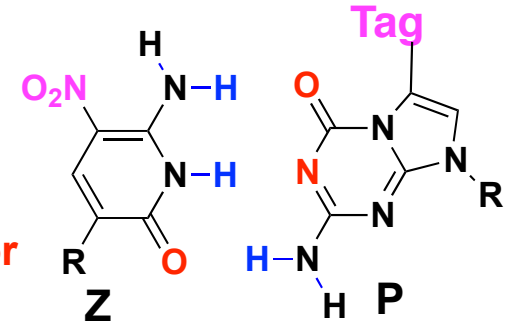
puADA
Acceptor
Donor
Acceptor

pyAAD
Acceptor
Acceptor
Donor



puDDA
Donor
Donor
Acceptor

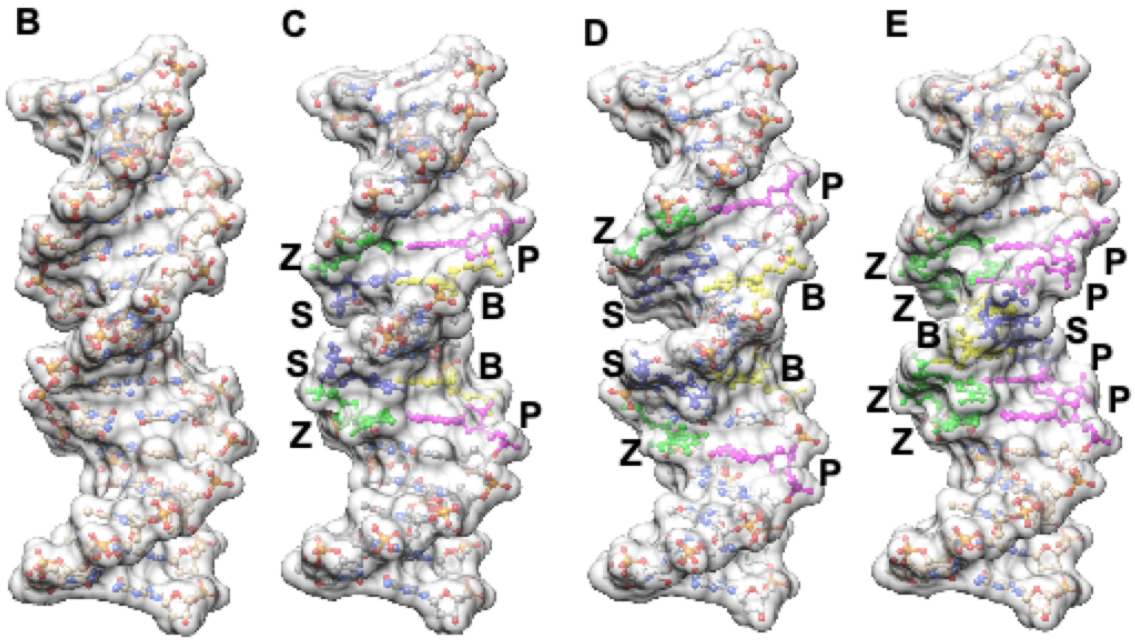
pyDDA
Donor
Donor
Acceptor



puAAD
Acceptor
Acceptor
Donor

Artificially Expanded Genetic Information System (AEGIS)

Synthetic alien DNA fits both Schrödinger and polyelectrolyte criteria

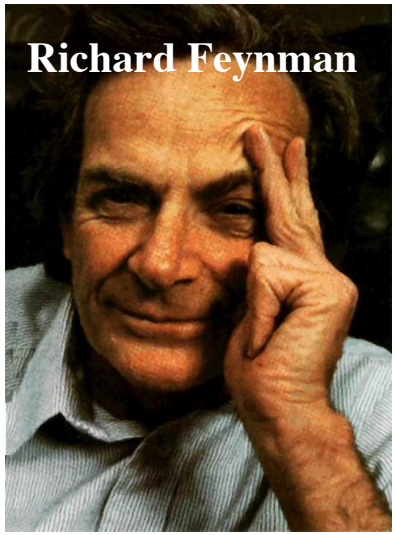


B) 8-letter hachimoji DNA **PB** (green), **P**C (red), **PP** (blue) atop GC DNA.
(**C**) 8-letter hachimoji DNA CTTAT**PB**TA**SZ**ATAAG (**PB**).
(**D**) 8-letter hachimoji DNA CTTA**P**C**B**TA**S**G**Z**TAAG (**PC**).
(**E**) 8-letter hachimoji DNA CTTAT**PPSBZZ**ATAAG (**PP**)

We are using synthesize to test our theories.

Imagine how much easier astronomy and geology would be if we could only synthesize our own stars, protosolar disks, and planets.

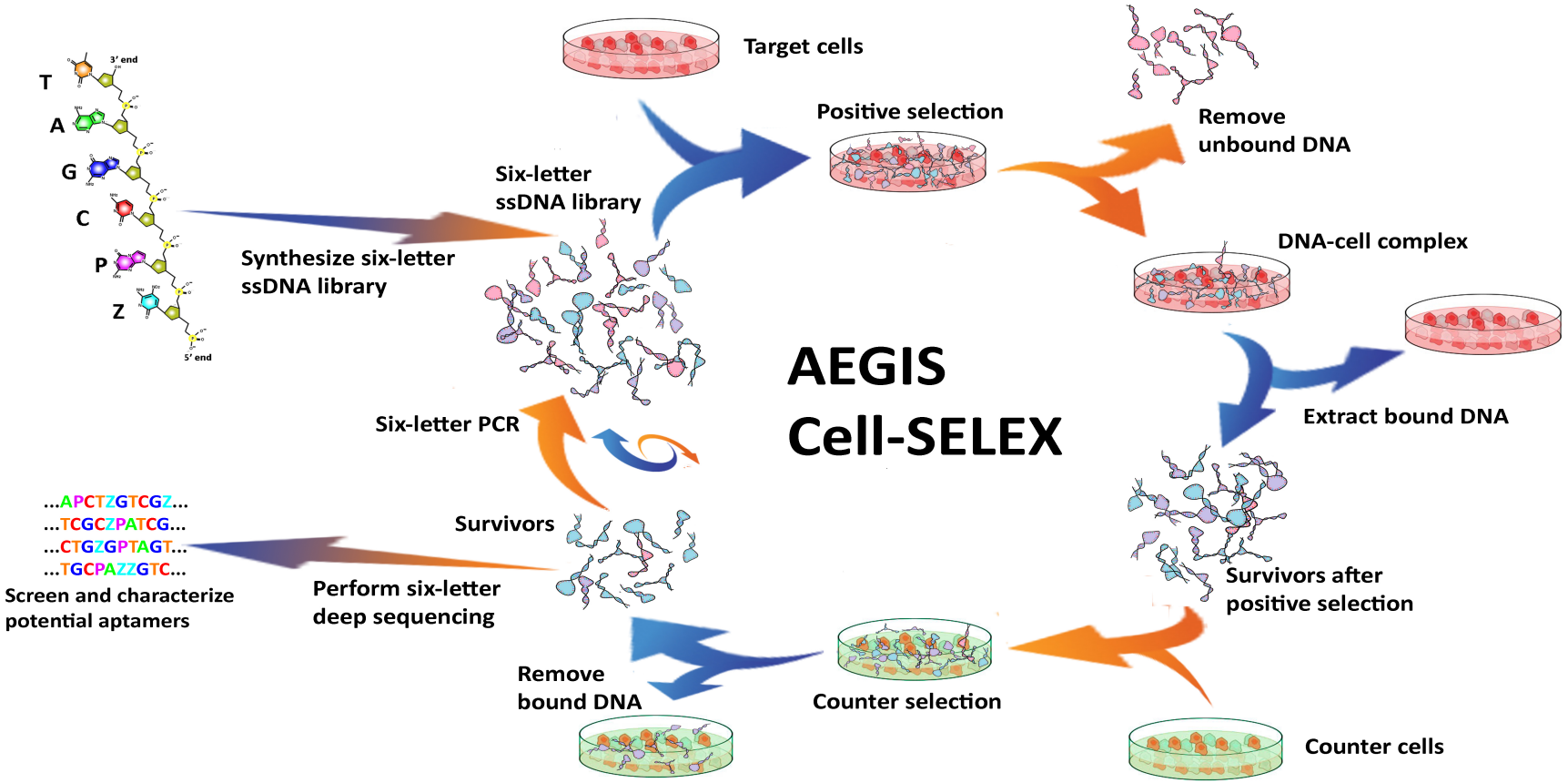
Feynman: "What I cannot synthesize, I do not understand"



AEGIS alien darwinian systems evolve in the lab to create functional molecules

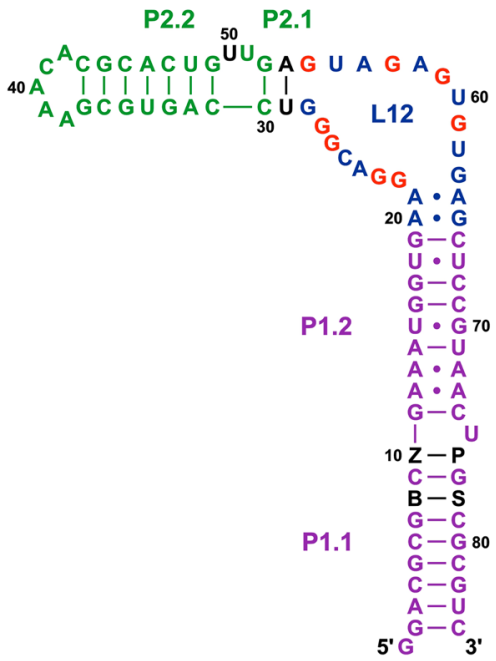
Some bind to cancer cells (liver, breast)
Some bind proteins (anthrax toxin, glypican 3)
Some catalyze reactions (ribonucleases)

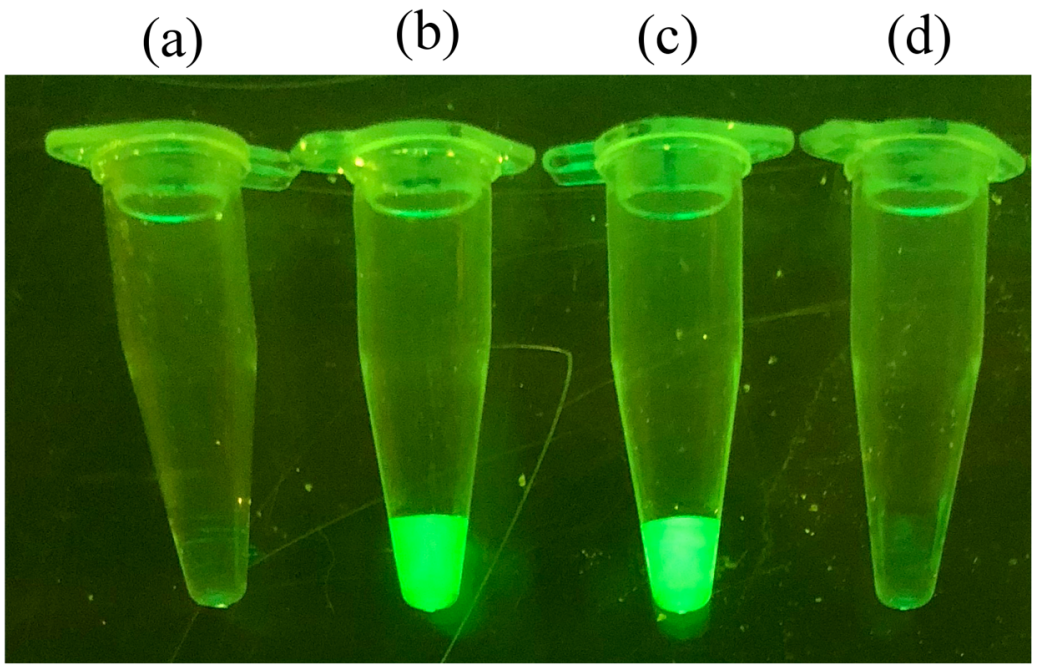
Zunyi Yang, Elisa Biondi
Liqin Zhang, Kwame Sefah, Weihong Tan

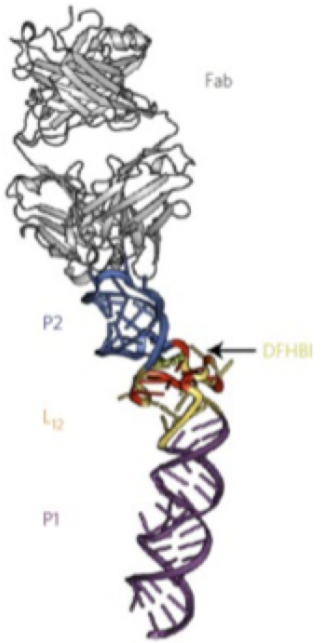


To understand biology, make some of your own

Some alien RNA binds fluor and glows



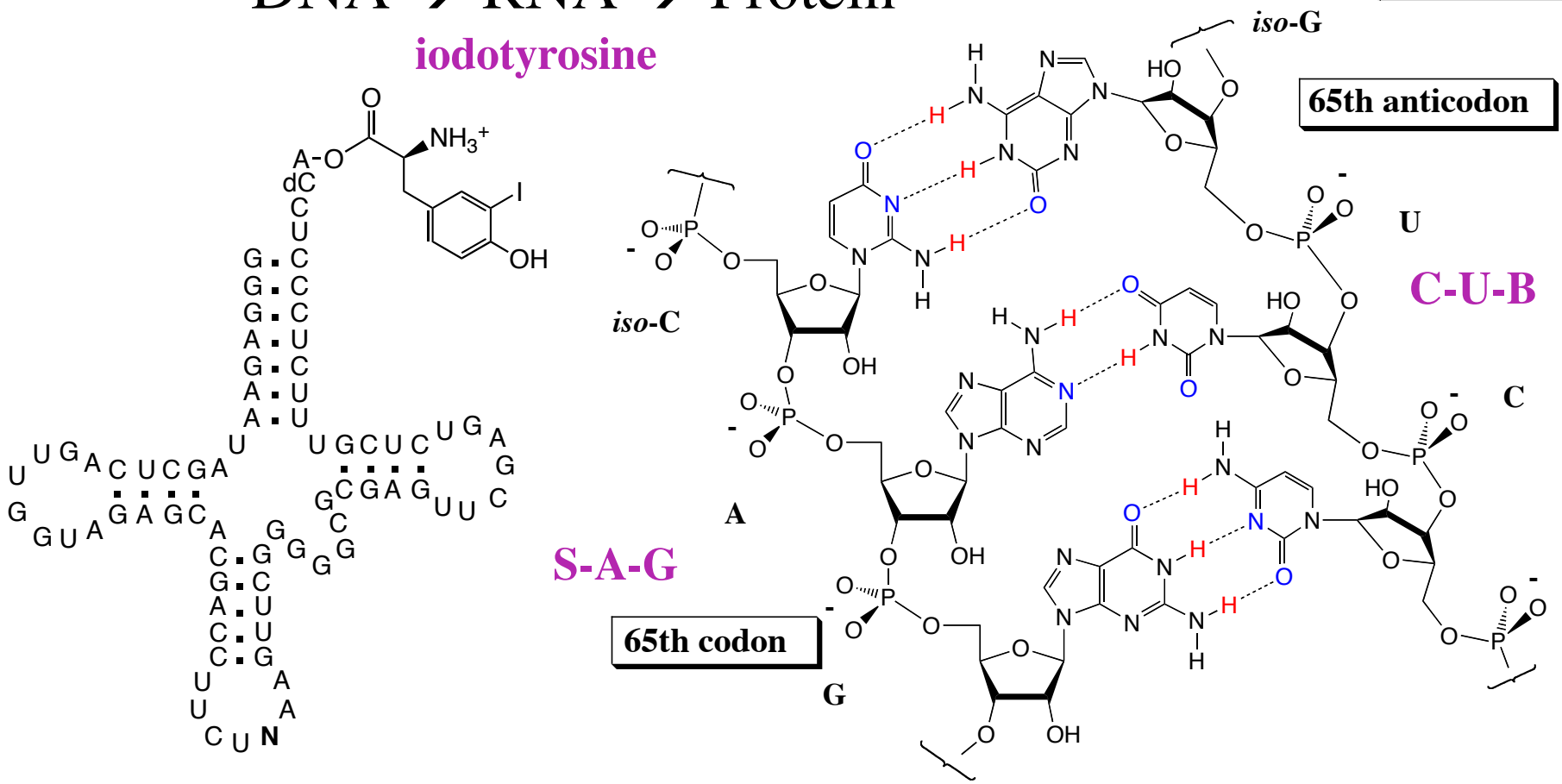




(a) Control with fluor only, lacking RNA,
(b) 8—Letter hachimoji RNA
(c) Native aptamer with fluor,
(d) fluor and aptamer with **Z** at position 50, replacing A:U pair at positions 53:29 with G:C to restore the triple. This places quenching **Z** chromophore near the fluor.

8-letter hachimoji encodes synthetic proteins

DNA → RNA → Protein



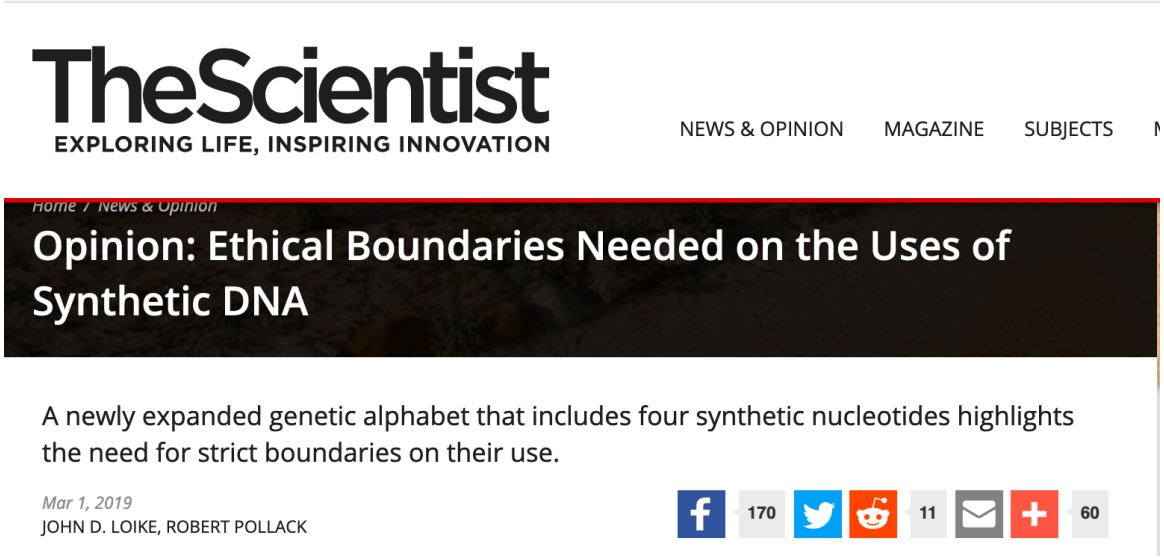
Bain, et al. (1992) Ribosome-mediated incorporation of non-standard amino acids into a peptide through expansion of the genetic code. *Nature* 356, 537-539

This is the 5'-SAG codon pairing with the 5'-CUB anticodon

Build what we value in biology on different platform=understanding

To be clear, we have *not* made artificial life

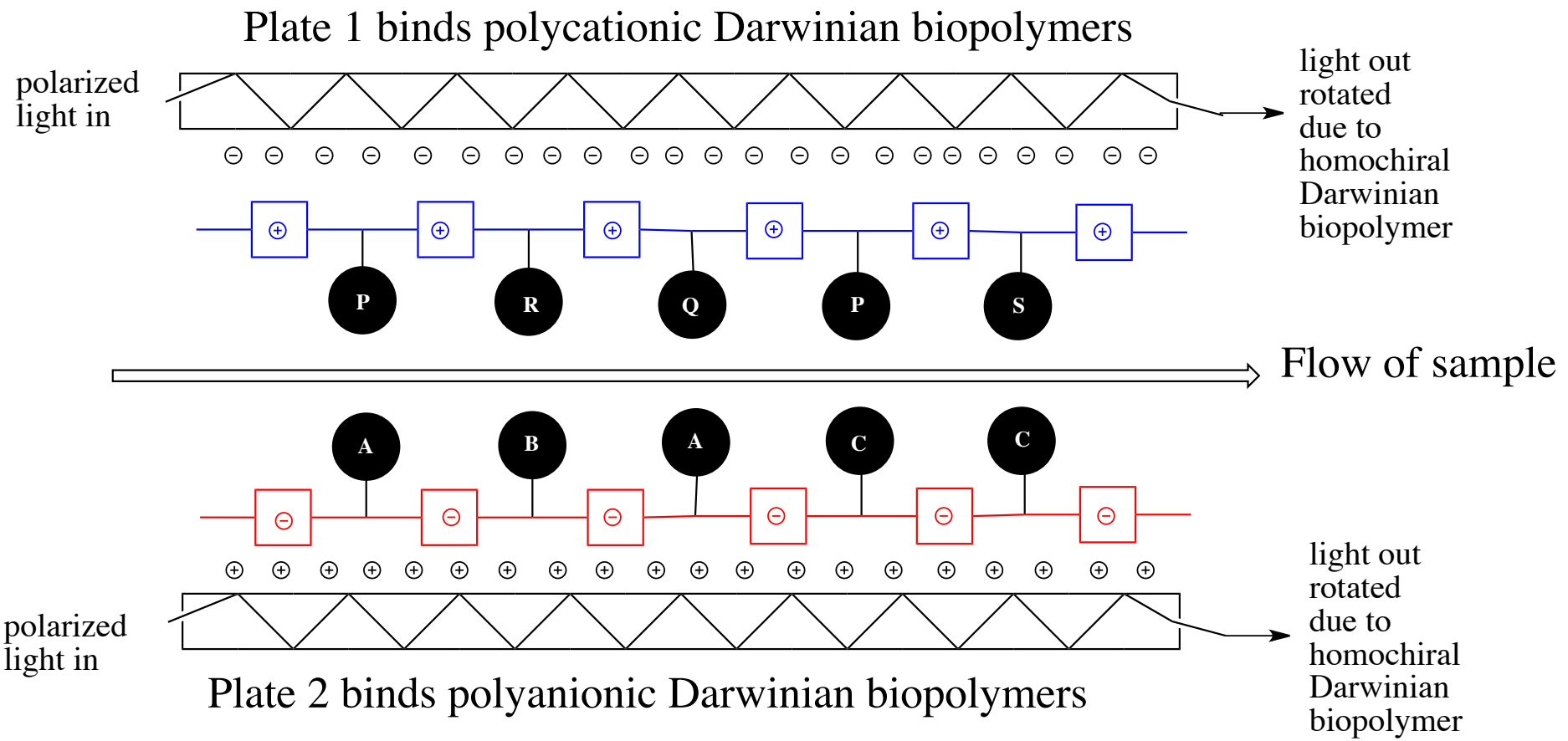
- Stores information with regular rules
- Information transferrable to other biopolymers
- Receiving system can have a selectable phenotype
- The biopolymer is able to evolve, fitting both the polyelectrolyte requirement and Schrödinger's aperiodic crystal structure
- ~~System must be self-sustaining, able to find its own food~~.



This omission was not sufficient to avoid criticism

Polyelectrolyte + Schrödinger criteria give a universal life detection strategy

Polyelectrolytes are easily concentrated from dilute solution by a support with many opposite charges.



This is what we should be using on Mars, Europa, & Enceladus

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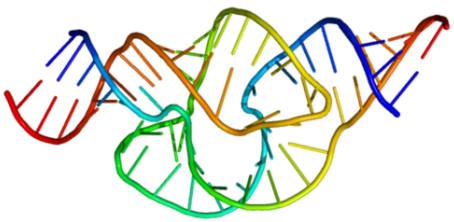
Hypothesis: Darwinism on Earth began with RNA as the Schrödinger polyelectrolyte

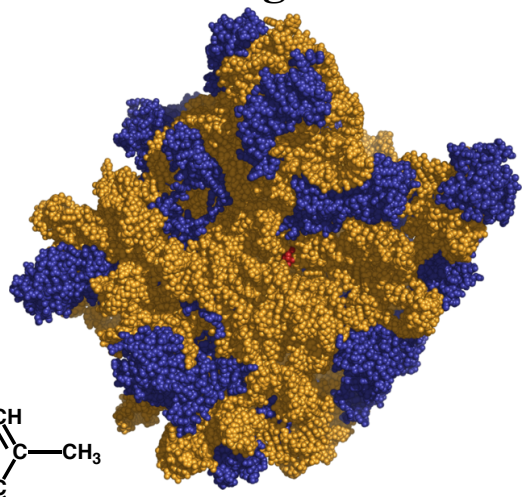
Darwinism began with **RNA** catalyzing template-directed synthesis of RNA, with replicable errors

Rich, A. (1962). On the problems of evolution & biochemical information transfer. in *Horizons In Biochemistry*, 103-126.

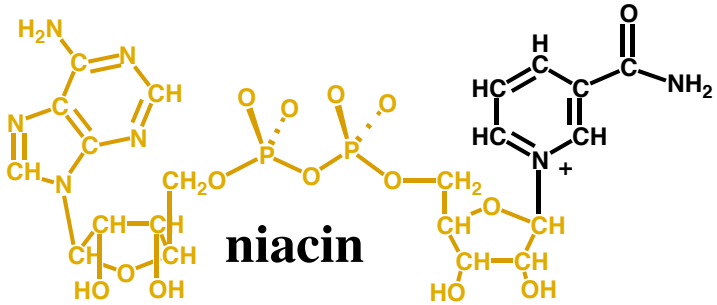


Ribosome, which *makes* **proteins**, is an **RNA** catalyst

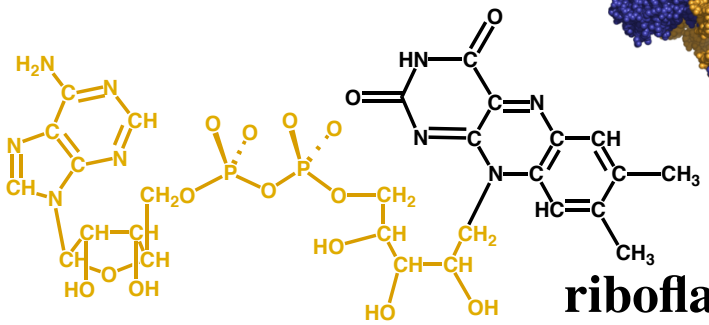
Lab example of Holliger et al.



DNA ← → messenger RNA → proteins, avoids chicken-or-egg problem (DNA or proteins first), & explains RNA cofactors central in your metabolism.



niacin



riboflavin

RNA combines information and performance in a single Darwinian biopolymer, not as well as AEGIS DNA.

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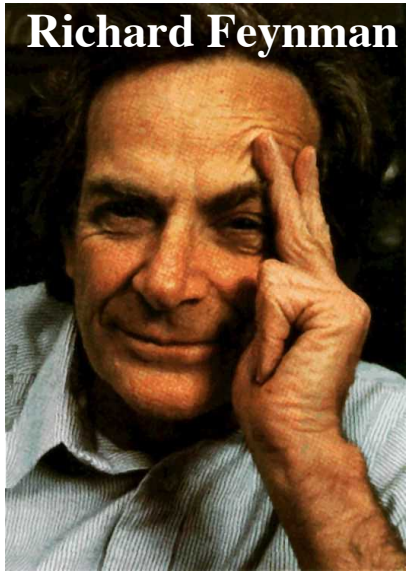
The problem with prebiotic chemistry

Intellectual discipline

Feynman: "People are easy to fool, and the easiest person to fool is yourself."

The field has few constraints, people love their own ideas, and few scientists credit data that contradict their ideas more than data that affirm their ideas.

The discipline of self-denial is hard to teach and apply.

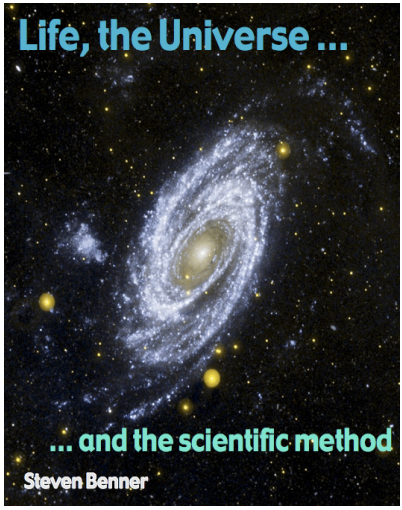


Focus on paradoxes is away to impose that discipline

"How wonderful that we have met with a paradox. Now we have some hope of making progress."

Niels Bohr

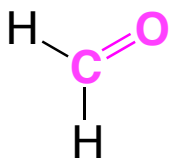
Otherwise, research is diffuse and focuses on culture-arising questions and historical contingency, whether or not they remain relevant to the big question.



"Settled science" says that ribose (the R in RNA) cannot be formed prebiotically

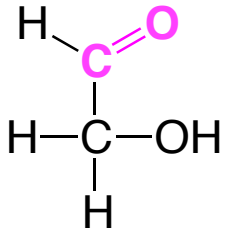
$C_nH_{2n}O_n$

Carbohydrates like **ribose** easily form tar. Known to *you*; Heat a bit, get carmel. Heat more, get brown stuff..



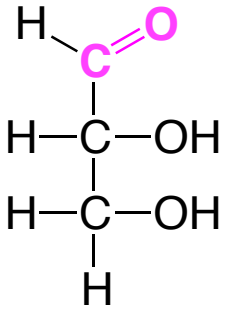
$C_1H_2O_1$

formaldehyde



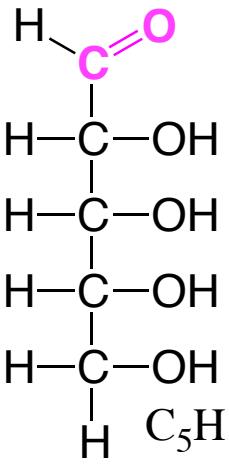
$C_2H_4O_2$

glycolaldehyde



$C_3H_6O_3$

glyceraldehyde



$C_5H_{10}O_5$

ribose

C=O group causes carmelization

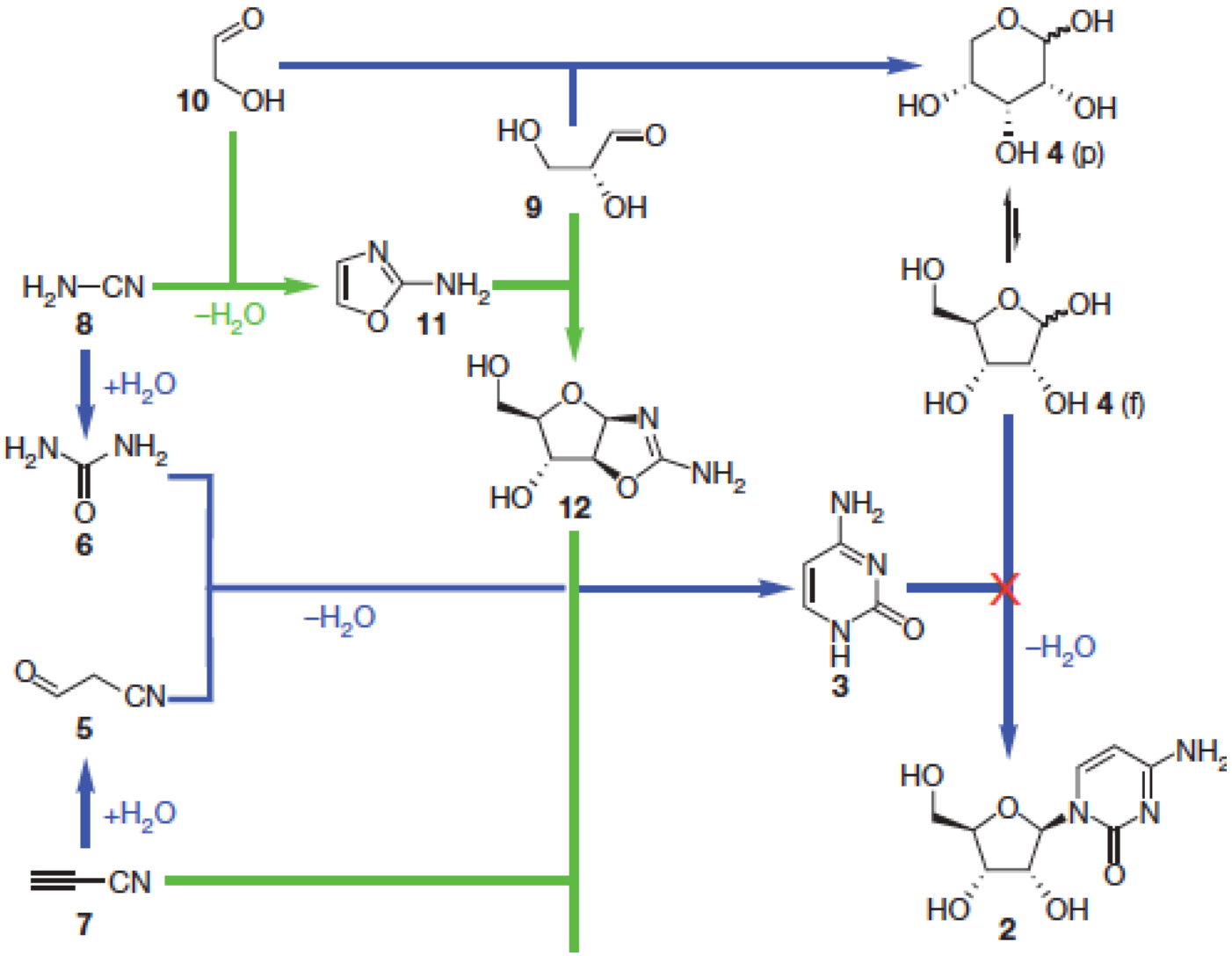
"stability considerations *preclude* ribose and other carbohydrates as prebiotic reagents …. Ribose and other sugars were not components of the first genetic material…" (Stanley Miller 1995)

Rhetoric is not an argument

Stanley Miller's experiment

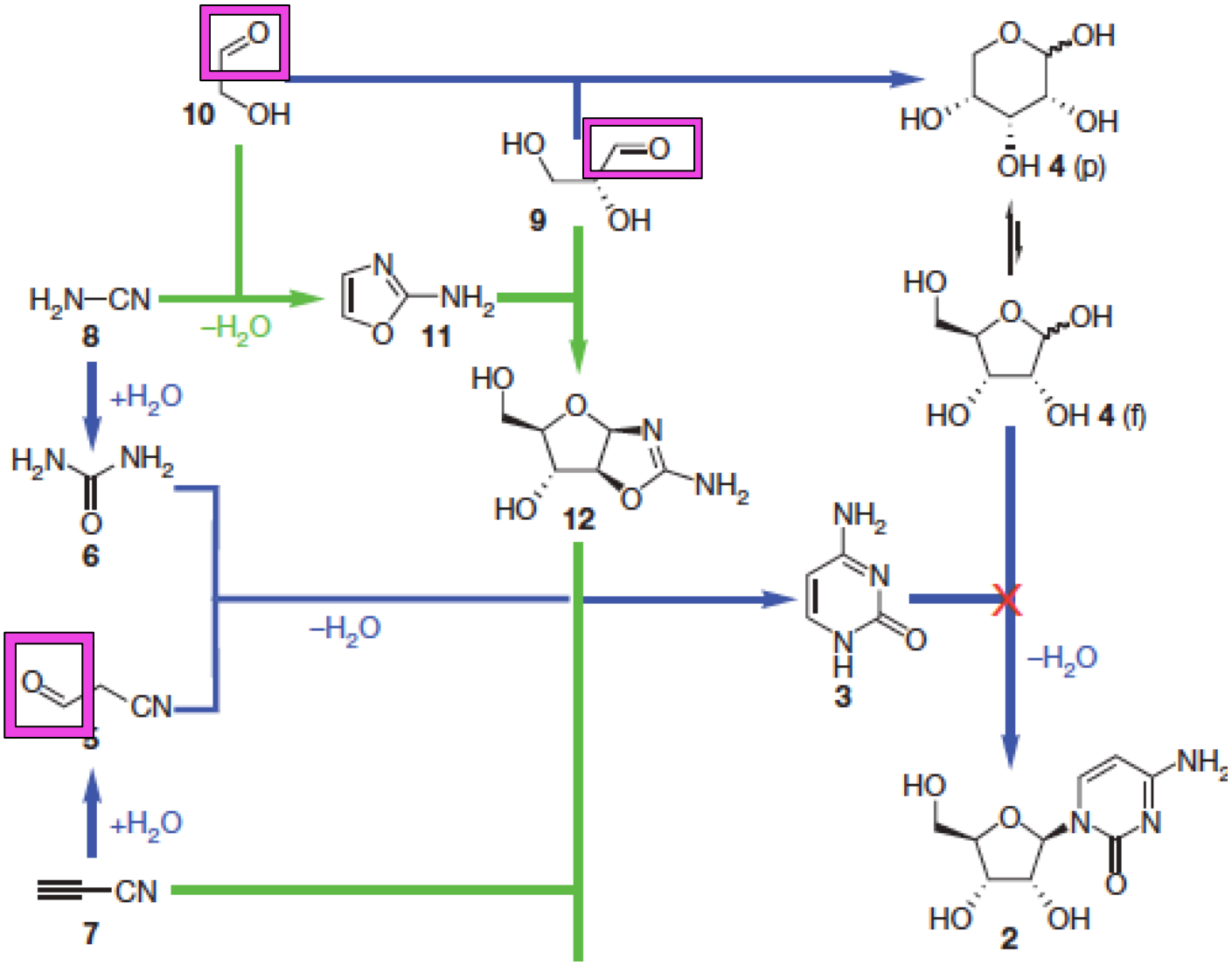
In fact, they digested with acid, used tungsten electrodes, got tar ...

Prebiotic carbohydrate C=O formation is invoked in many paths to RNA building blocks



For example, Powner *et al.* (2009) *Nature* **459**, 239–242. **Can you find the unstable carbohydrates?**

Prebiotic carbohydrate C=O formation is invoked in many paths to RNA building blocks



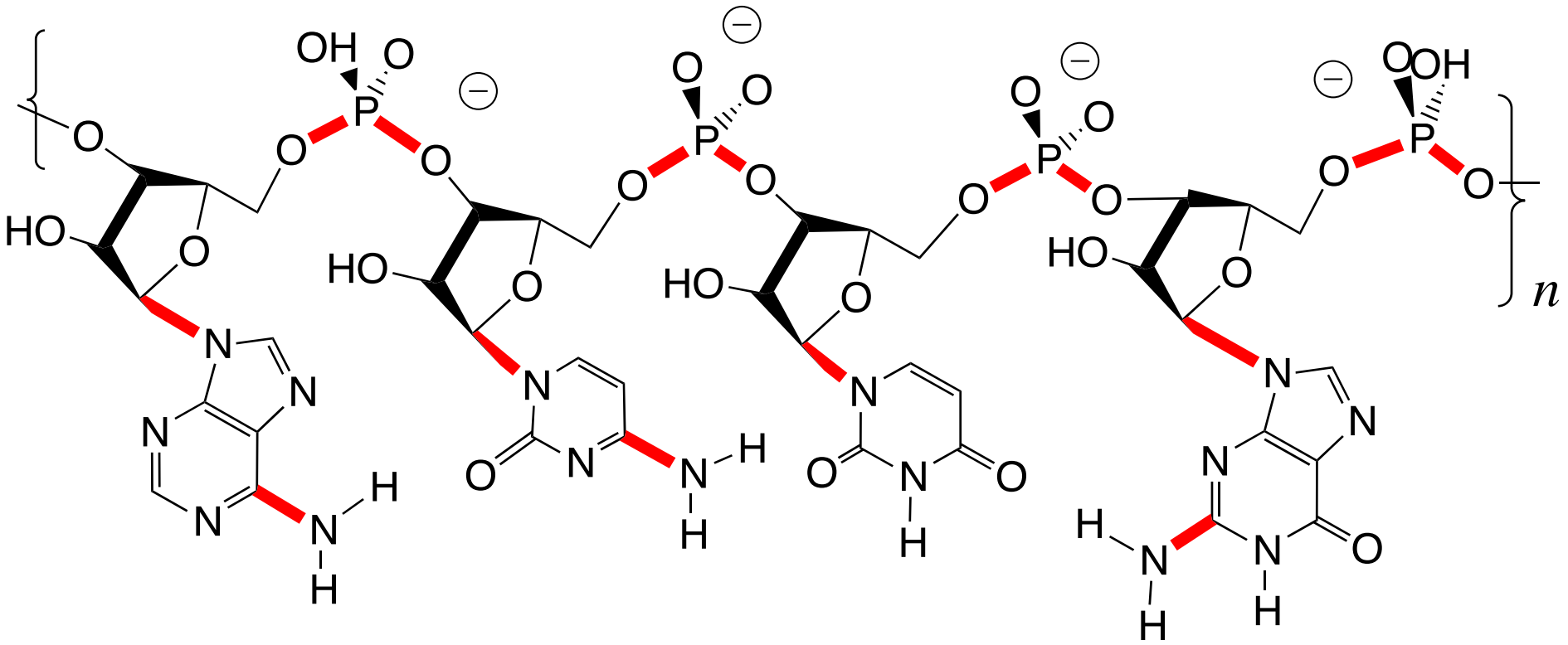
For example, Powner *et al.* (2009) *Nature* 459, 239–242. Can you find the unstable carbohydrates?

Look for C=O

If C=O molecules are needed in accumulated amounts, more explaining is needed.

"Settled science" says that even if you form ribose and RNA from it, RNA falls apart in water.

Every bond in red is thermodynamically unstable in water



The paradox constrains project selection. If ribose cannot be made prebiotically, and even if made, bonds to have it to form RNA cannot be made in water, and even they can, RNA falls apart in water, we focus here to research the RNA-first model for the origin of terran Darwinism.

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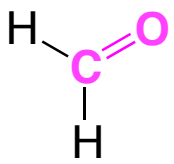
Looking for premises that you forgot to include in your logic

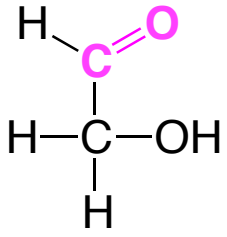
$C_nH_{2n}O_n$

Carbohydrates like **ribose** easily form tar. Known to *you*; Heat a bit, get carmel. Heat more, get brown stuff..



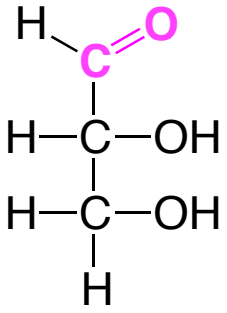
$C_1H_2O_1$

formaldehyde



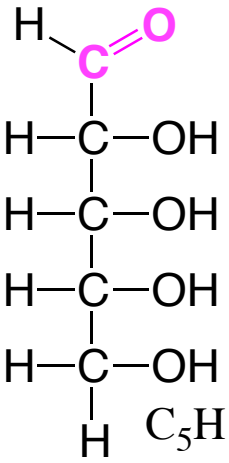
$C_2H_4O_2$

glycolaldehyde



$C_3H_6O_3$

glyceraldehyde



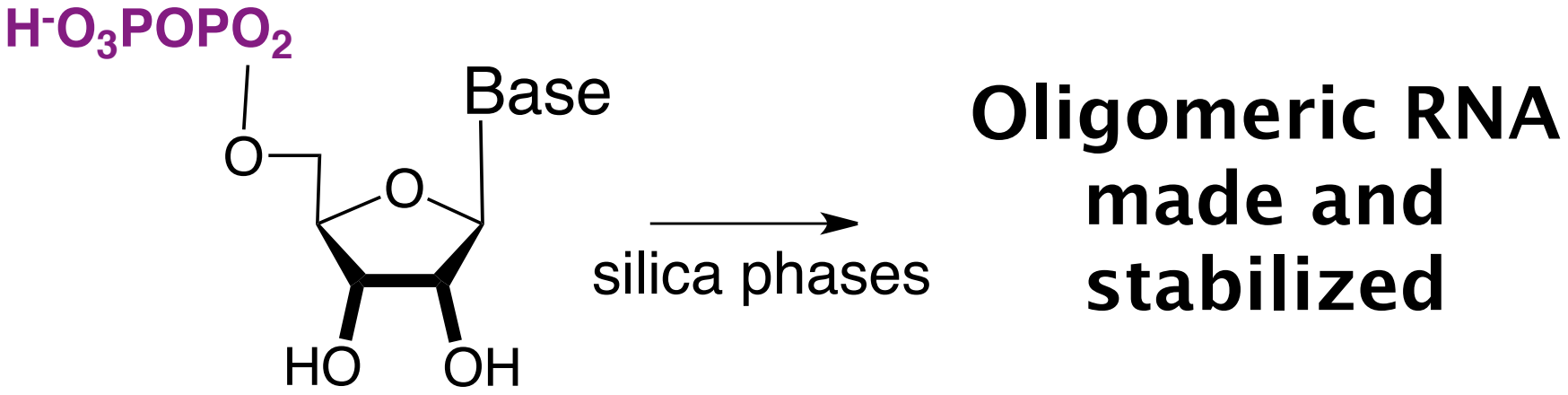
$C_5H_{10}O_5$

ribose

But your kitchen experiment did not put these things in rocks

Let us work backwards, summarizing results.

RNA is formed on silica phases from RNA nucleoside diphosphates



Biondi, Howell, Benner (2017) Opal absorbs and stabilizes RNA. A hierarchy of prebiotic silica minerals. *Synlett*, **28**, 84-88.
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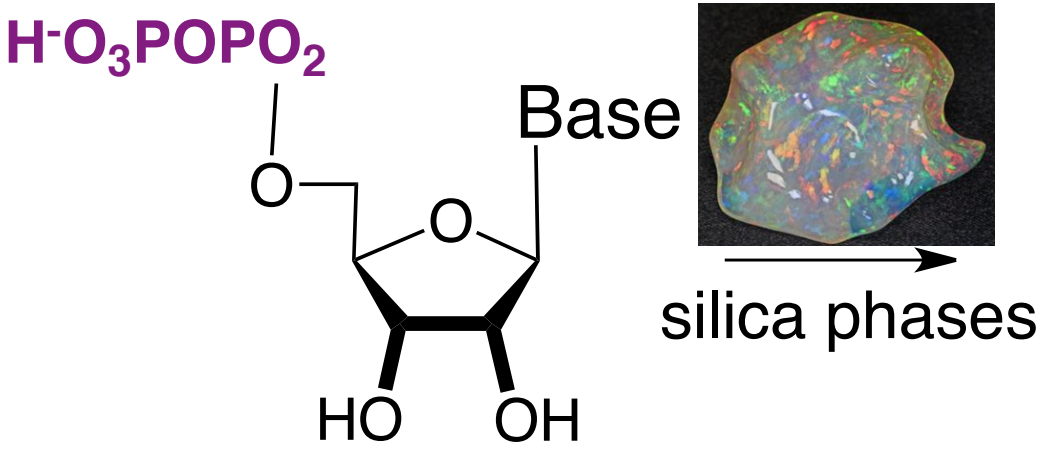
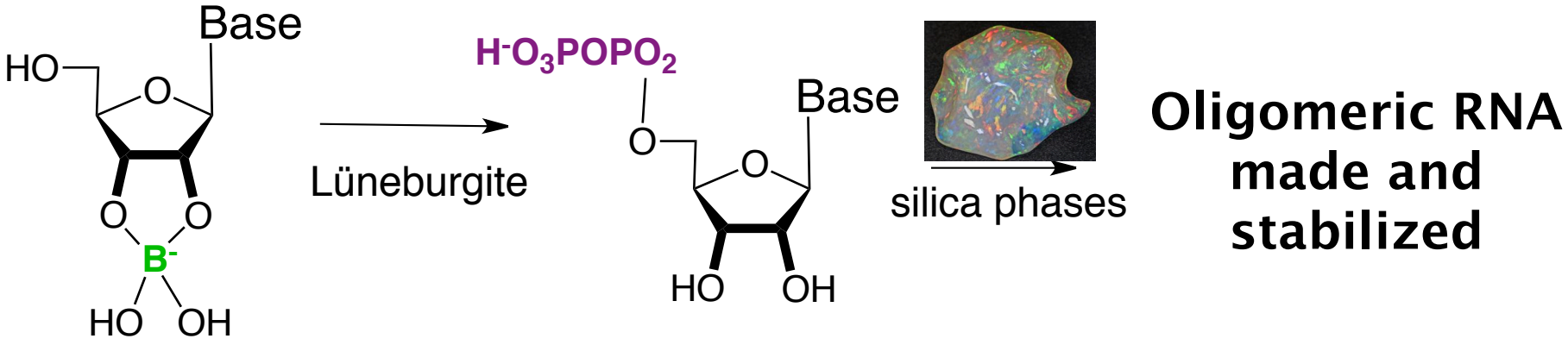


Oligomeric RNA made and stabilized

Biondi, Howell, Benner (2017) Opal absorbs and stabilizes RNA. A hierarchy of prebiotic silica minerals. *Synlett*, **28**, 84-88.
Biondi, Furukawa, Kawai, Benner (2017) Adsorption of RNA on mineral surfaces and precipitates. *Beilstein J. Org. Chem.* **13**, 393.

But where do the ribonucleoside diphosphates come from?

Magnesium borophosphate mineral lüneburgite make diphosphates from nucleosides



Kim, Furukawa, Kakegawa, Bita, Scorei, Benner (2016) Evaporite borate-containing mineral ensembles make phosphate available and regiospecifically phosphorylate ribonucleosides: Borate as a multifaceted problem solver in prebiotic chemistry. *Angew. Chem. Int. Ed.* **55**, 15816-15820,

This also offers a solution to the "phosphate paradox", which arises from the low solubility of calcium phosphate minerals, and a part of the "tar paradox"; **borate** controls where the **phosphate** goes.

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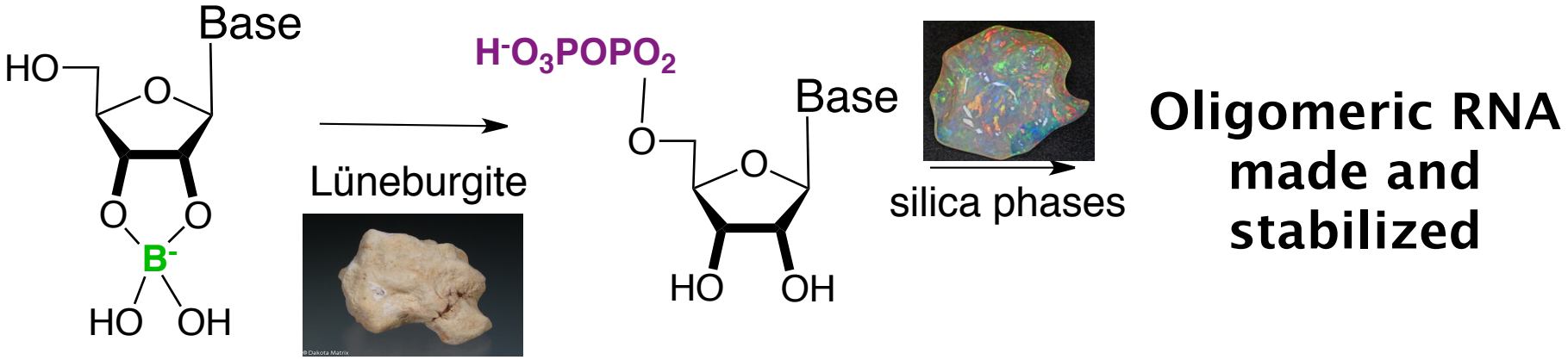

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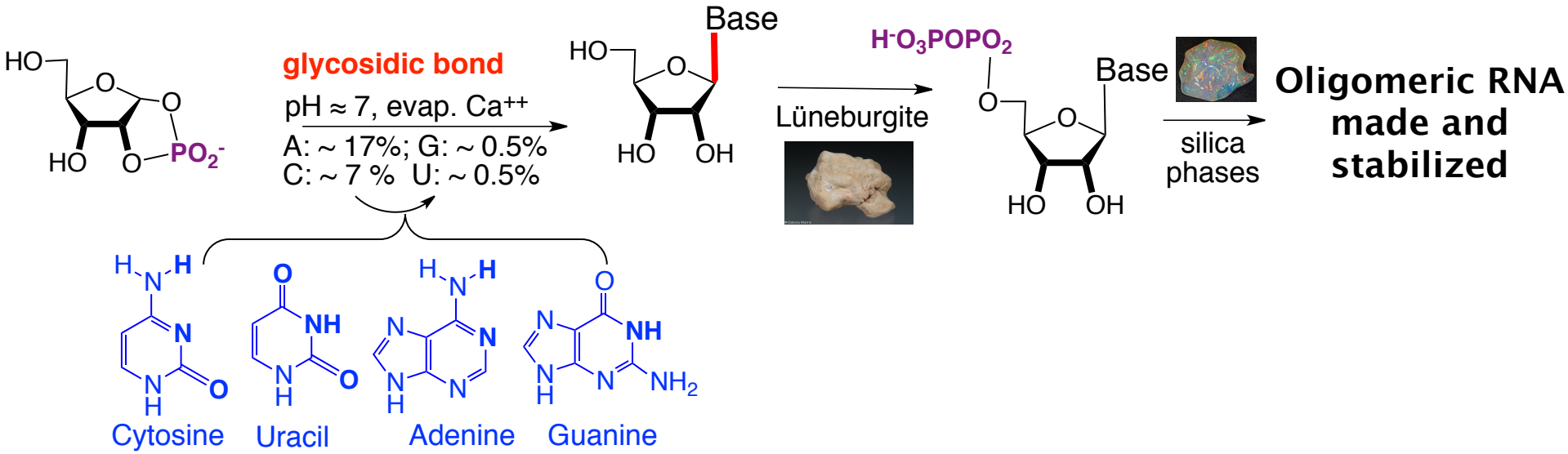
But where do the ribonucleosides come from?

Evaporation makes nucleosides from ribose cyclic phosphate

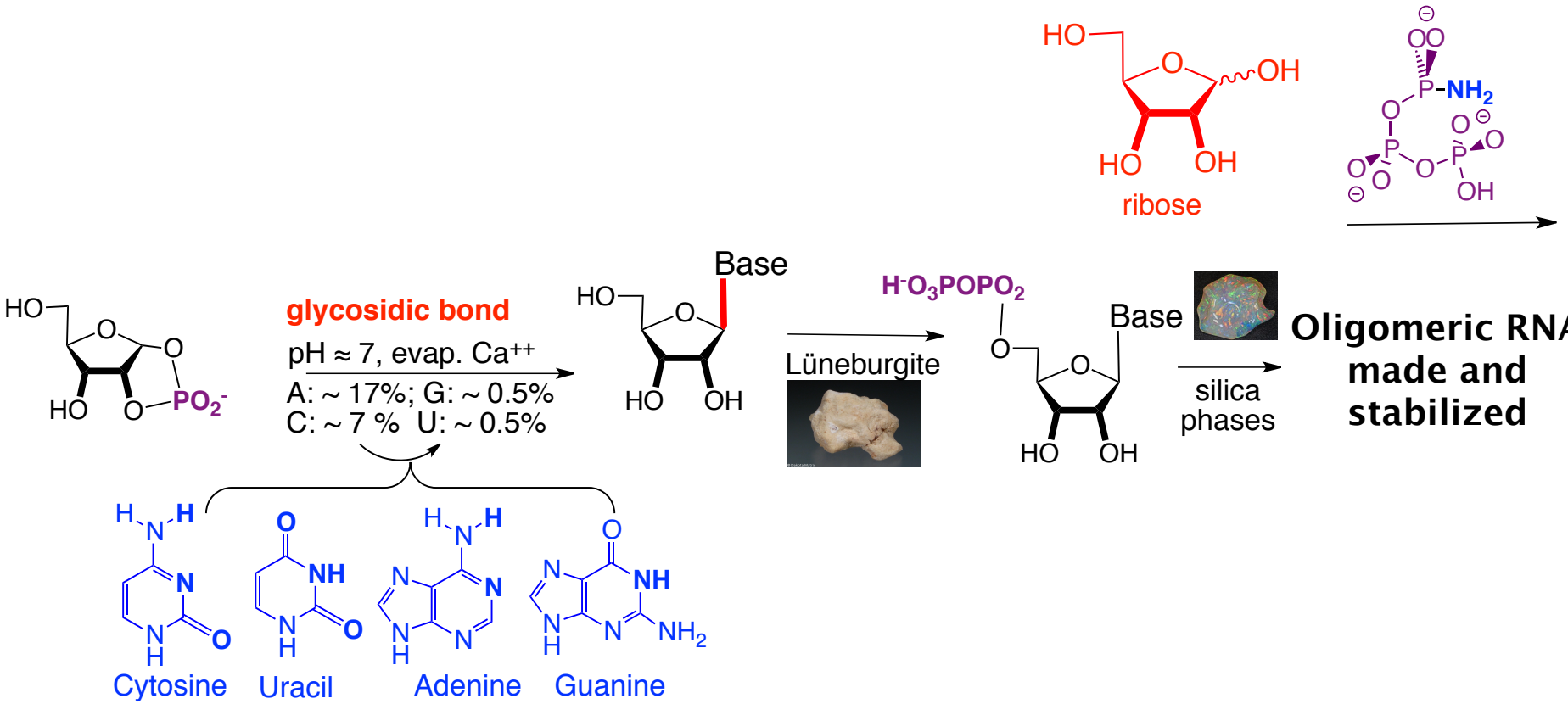


Kim, Kim (2019). A Prebiotic Synthesis of Canonical Pyrimidine and Purine Ribonucleotides. *Astrobiology*, *19*(5), 669-674.

This also offers a solution to part of the "water paradox", which arises from the instability of the **glycosidic** bond in water.

But where do the ribose cyclic phosphates come from?

Reaction of ribose with amides from cyclic trimetaphosphate gives ribose cyclic phosphates



Krishnamurthy, Guntha, Eschenmoser (2000) Regioselective α-phosphorylation of aldoses in aqueous solution. *Angew. Chem. Int. Ed.* **39**, 2281-2285.

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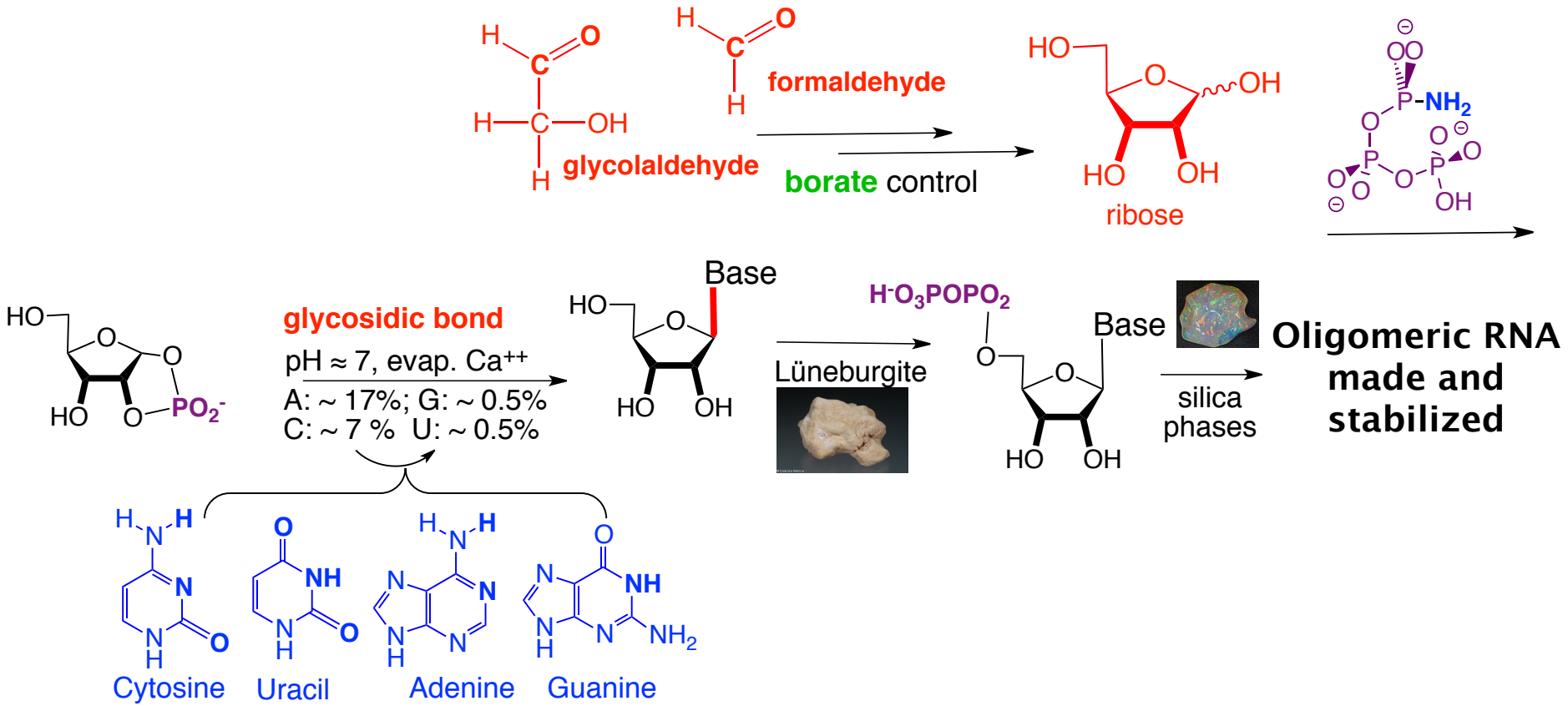
But where does the ribose come from?

In the presence of borate, pentoses like ribose come from formaldehyde and glycolaldehyde

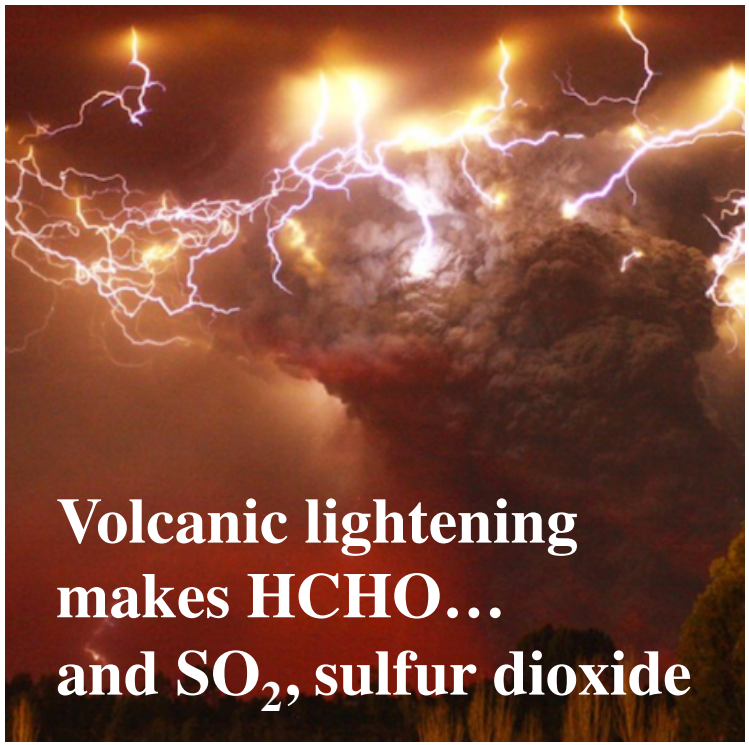


Kim,Ricardo,Illangkoon,Kim,Carrigan,Frye,Benner (2011) Synthesis of carbohydrates in mineral-guided prebiotic cycles. *J. Am. Chem. Soc.* **133**, 9457-9468.

This also offers a solution to part of the "tar paradox", where **borate** controls the carbohydrate carmelization that we spoke of.

But where did the carbohydrate reservoirs come from?

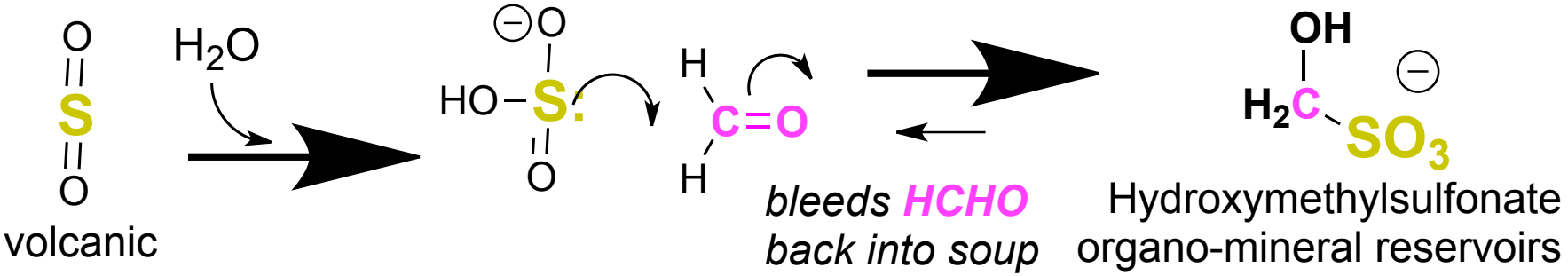
Sulfur dioxide outgasses from mantle at the redox state studied by Dustin



SO_2 reacts with carbohydrate C=O to reversibly form stable sulfonates. These are mineral reservoirs of a carbohydrates.

"Bespoke chemistry", certain to have happened on Hadean Earth.

Kawai, McLendon, Kim, Benner (2019) Hydroxymethanesulfonate from volcanic SO_2. A mineral reservoir for formaldehyde and other simple carbohydrates in prebiotic chemistry. *Astrobio*. **19**, 506.



The sulfonates accumulate on dry land in tons

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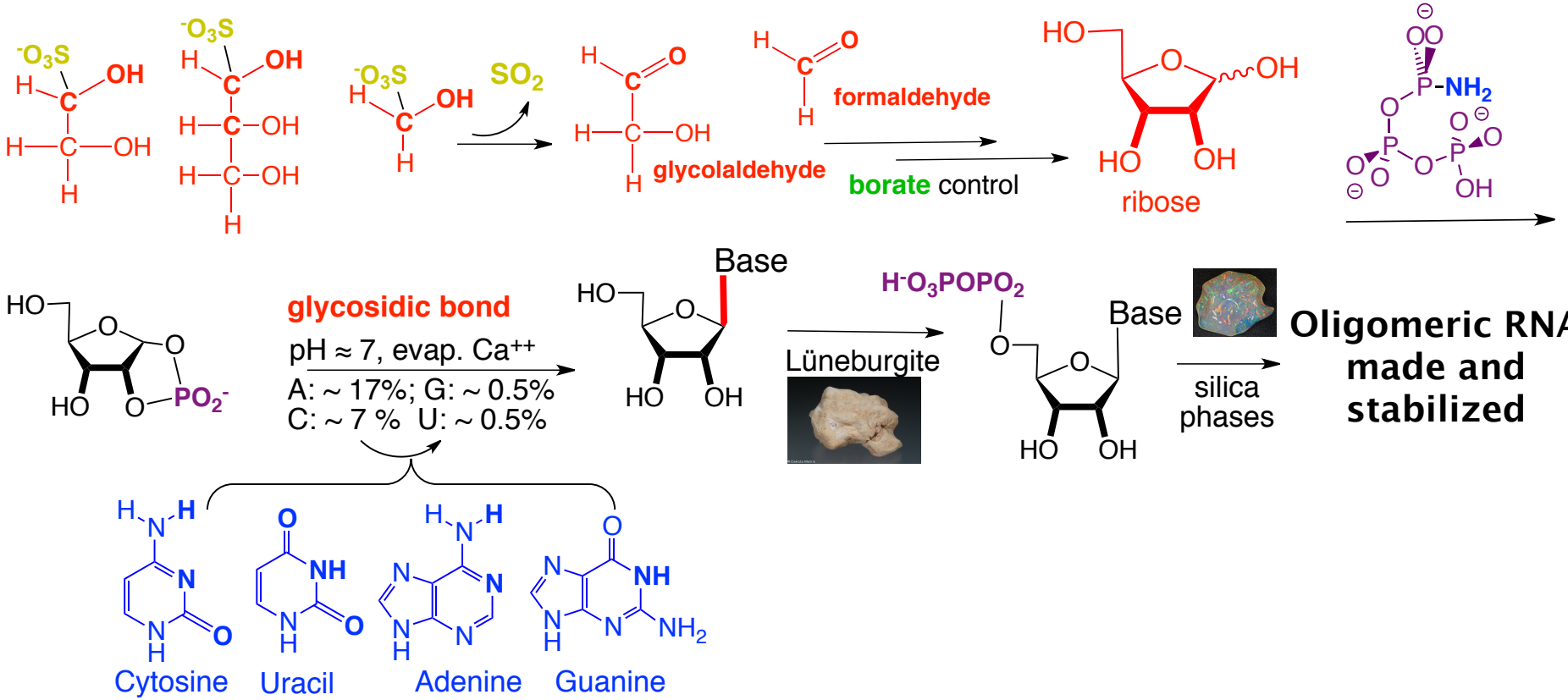

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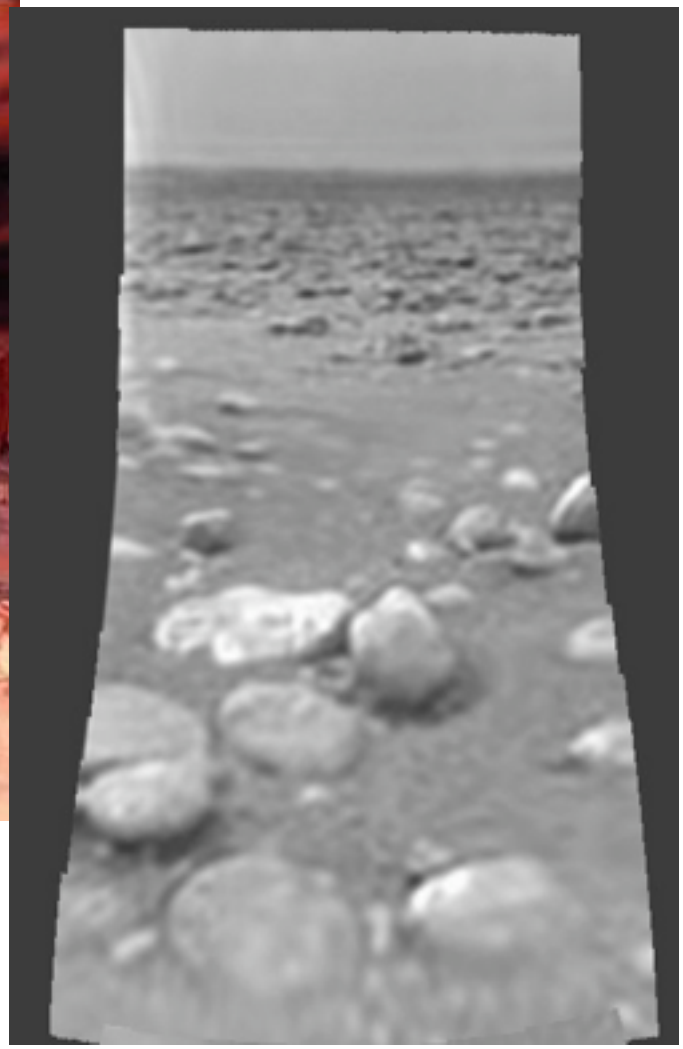
Analogy is surface of modern Titan

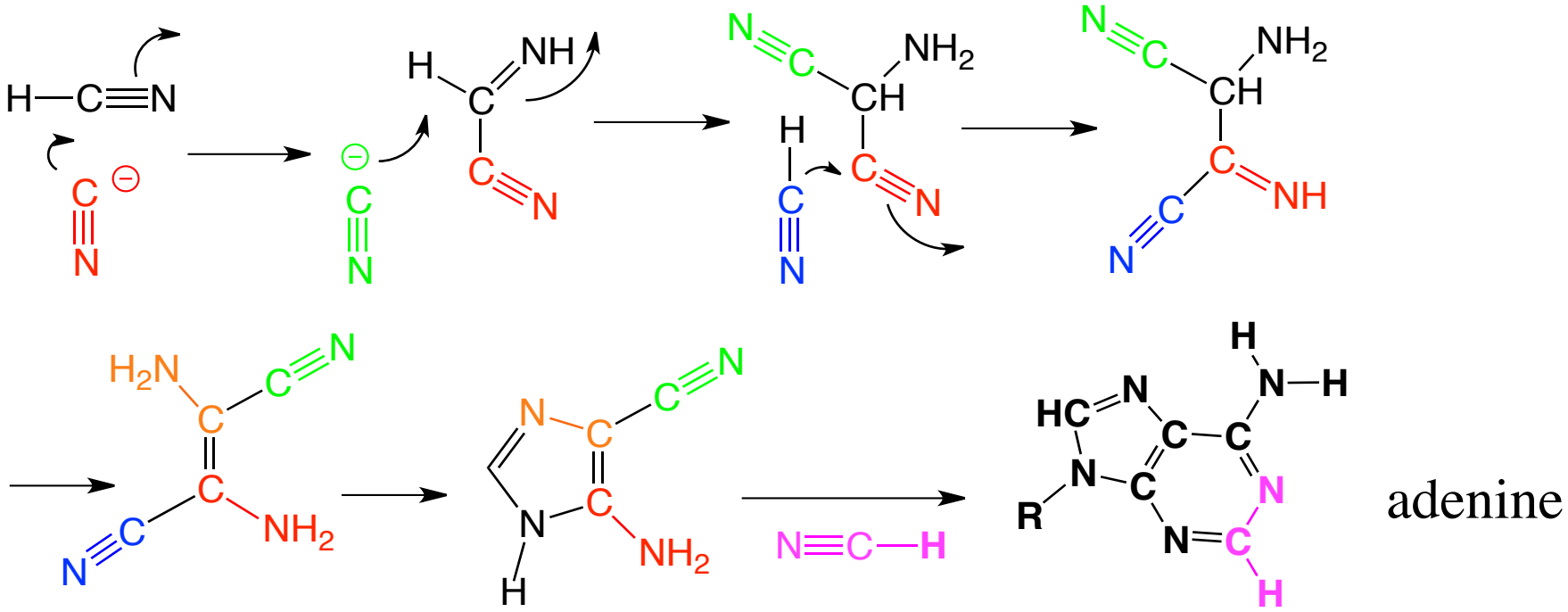
Metric tons of accumulated organics on dry land



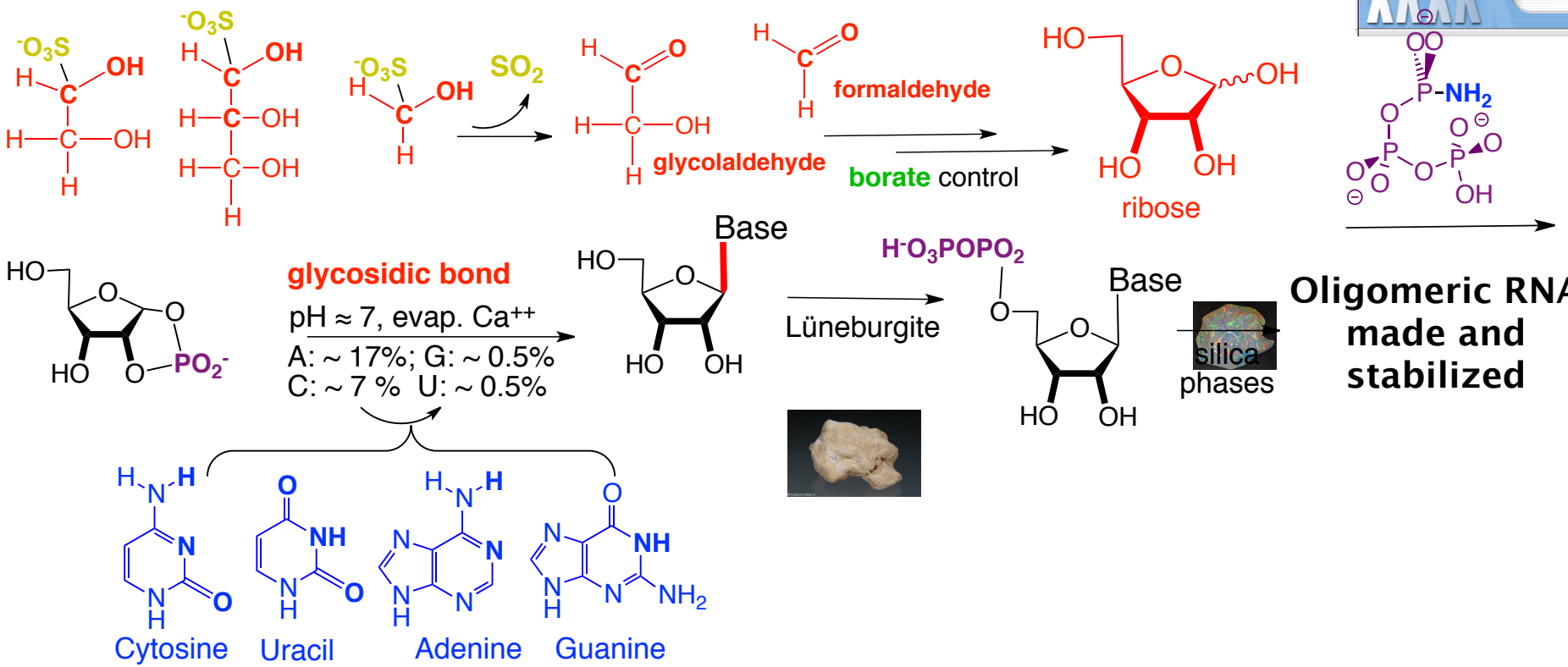
New problems and paradoxes

Were borate and phosphate minerals available
Was there dry land?

But another **paradox**. **If the atmosphere had the oxidation state of SO_2, it could *not have formed* HCN, HNCNH, HCCCN, and other reduced organics (N = NH_3). These are needed to form the nucleobases.**



I *assumed* that nucleobases were available.



Benner, Bell, Biondi, Brasser, Carell, Kim, Mojzsis, Omran, Pasek, Trail (2019) When did life likely emerge on Earth in an RNA-first process? *Angew. Chem.* Submitted.

Focusing on this paradox was quite interesting ...

... Emerging from a workshop held at Georgia Tech (Oct. 2018) sponsored by the John Templeton Foundation.

Service (2019) Seeing the dawn. Evidence lines up to offer a new view of how life on our planet may have emerged. *Science* **363** (6423) 116-119

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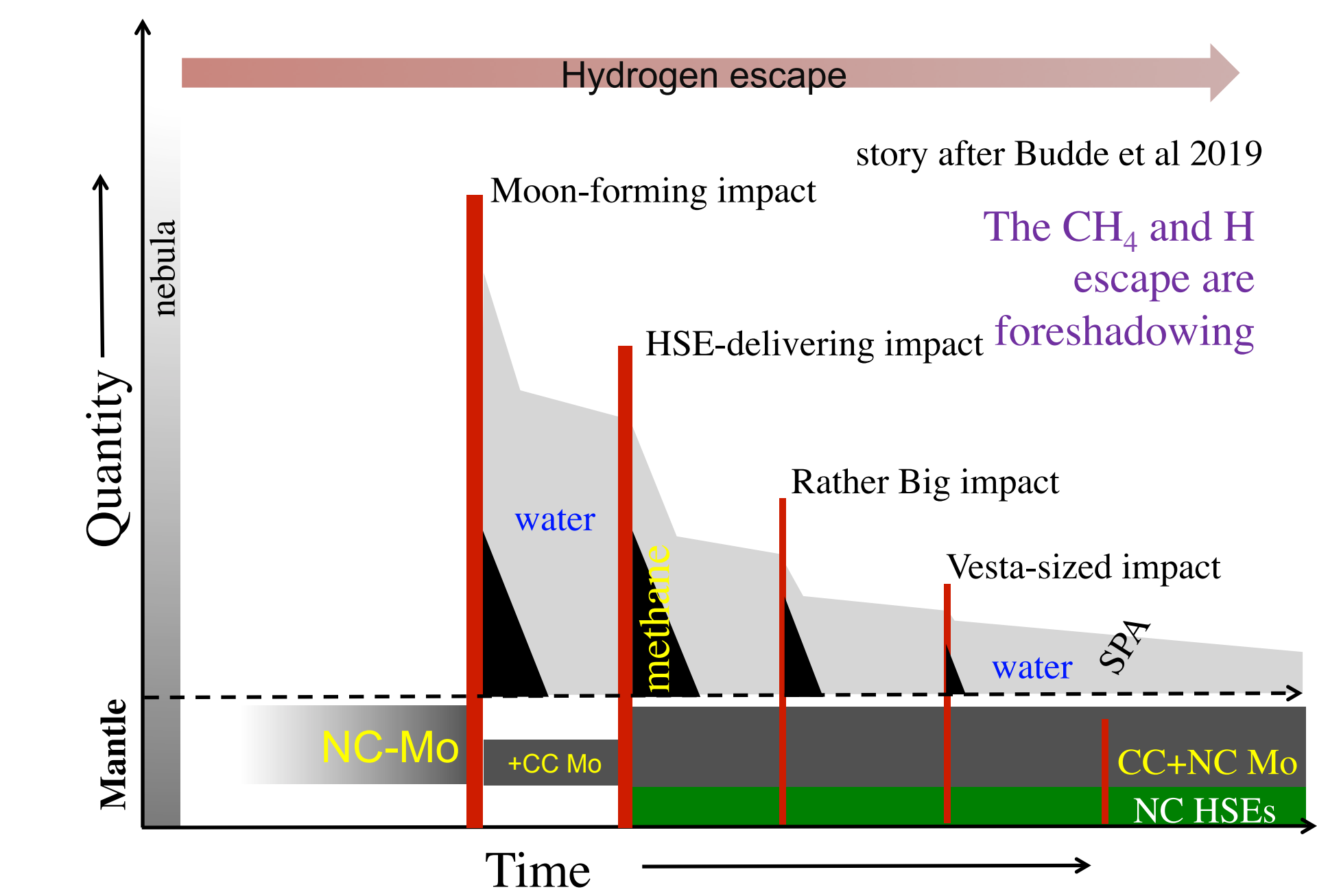
Earth's crust has a veneer of iron-loving elements delivered after the core was closed.

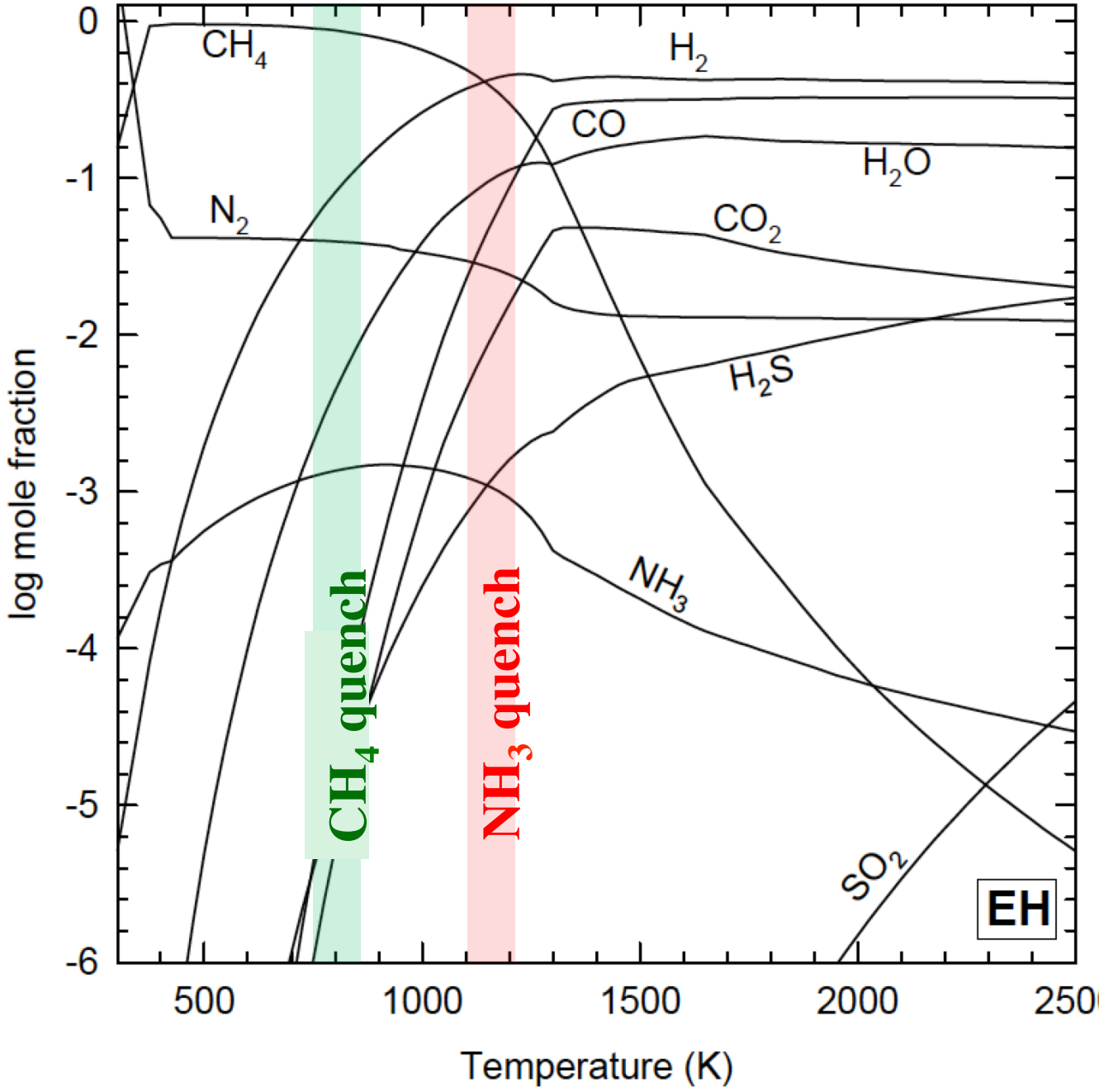
Were these "siderophiles" delivered in one large impactor, many small impactors?

The Moon/Earth veneer ratio is lower than the ratio of gravitational cross-sections, implying a single impactor.
An impactor large enough to account for all of the veneer would be about the size of the Moon.
It would have its own iron core.
Unless it hit square-on, the core would fragment, delivering large amounts of iron to the atmosphere, and create a reducing atmosphere productive for HCN, NH_3, HNCNH, HCCCN and hence, nucleobases.

Mojzsis, Brasser, Kelly, Abramov, Werner (2019) Onset of giant planet migration before 4480 million years ago." *arXiv:1903.08825*.



Thanks to Kevin Zahnle



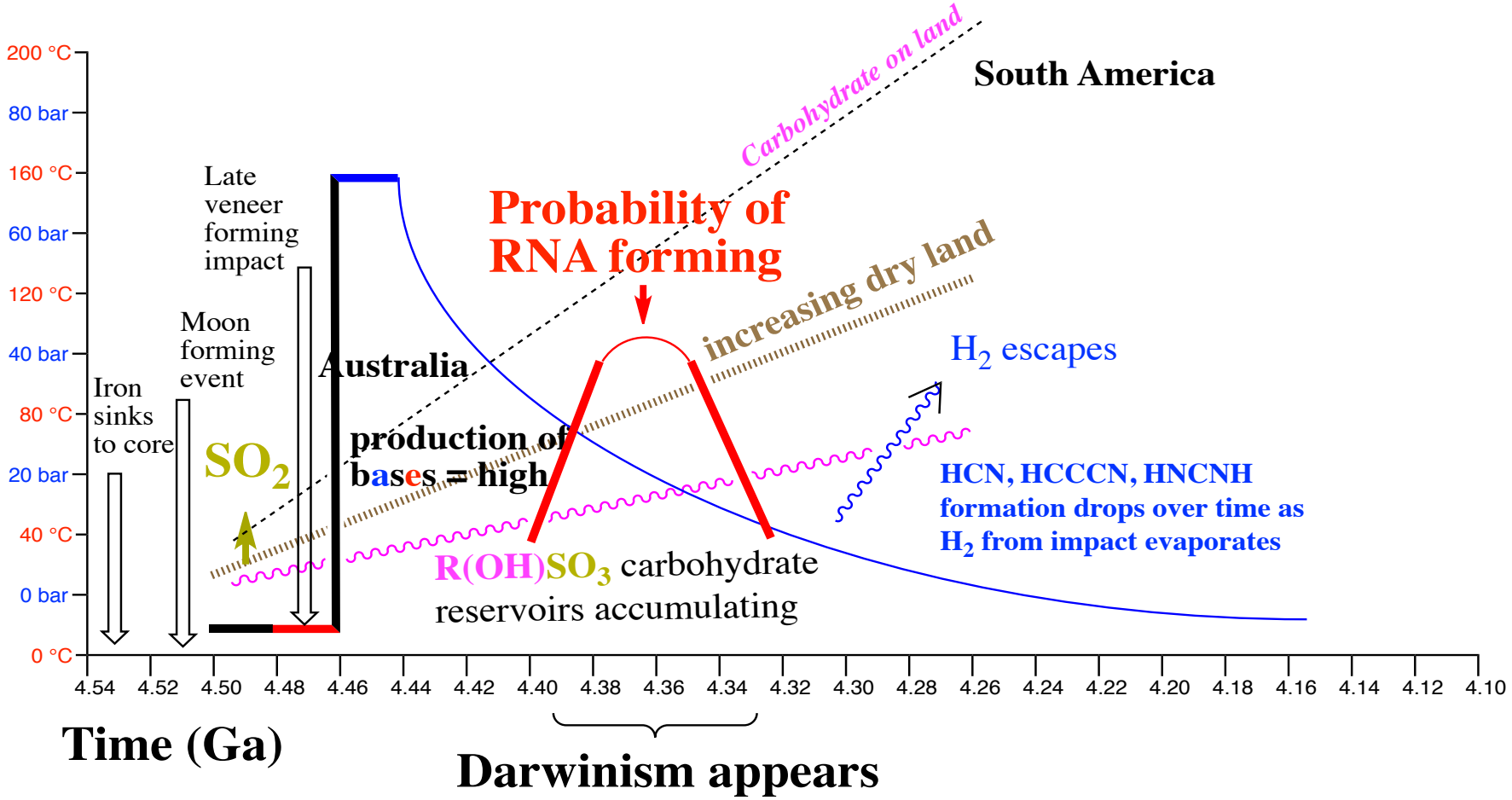
Schaefer and Fegley 2009

Gases equilibrated to Enstatite Chondrites

My quench temperatures

The QFI buffer at 100 bars is **Very** favorable for CH_4 and NH_3

If veneer-delivering impact was last to sterilize, and if it occurred at ~4.47 Ga, then optimum for RNA formation is 4350 ± 50 MY



Now, we are not used to ± 50 MY precision at this antiquity. So this estimate has met some ridicule.

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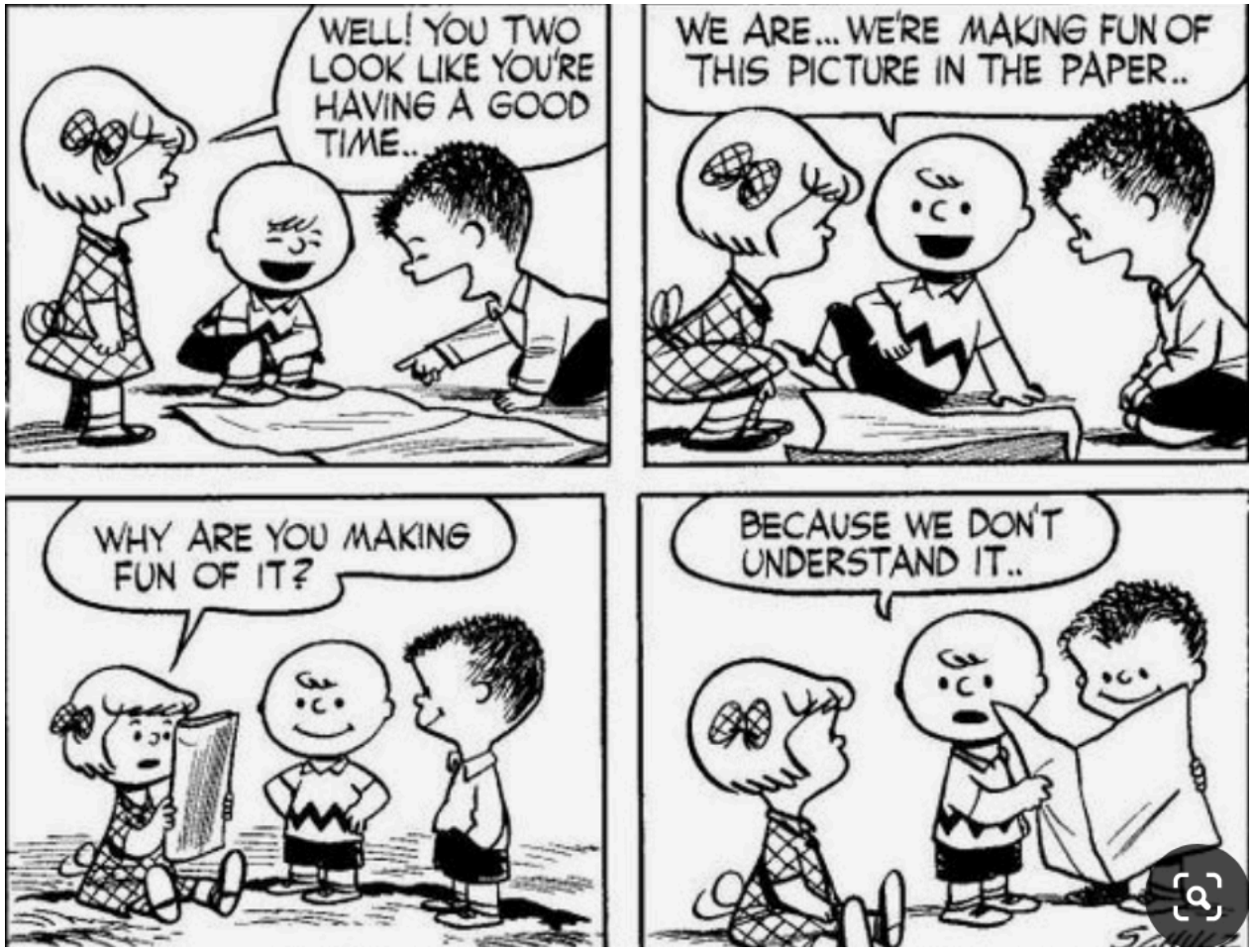
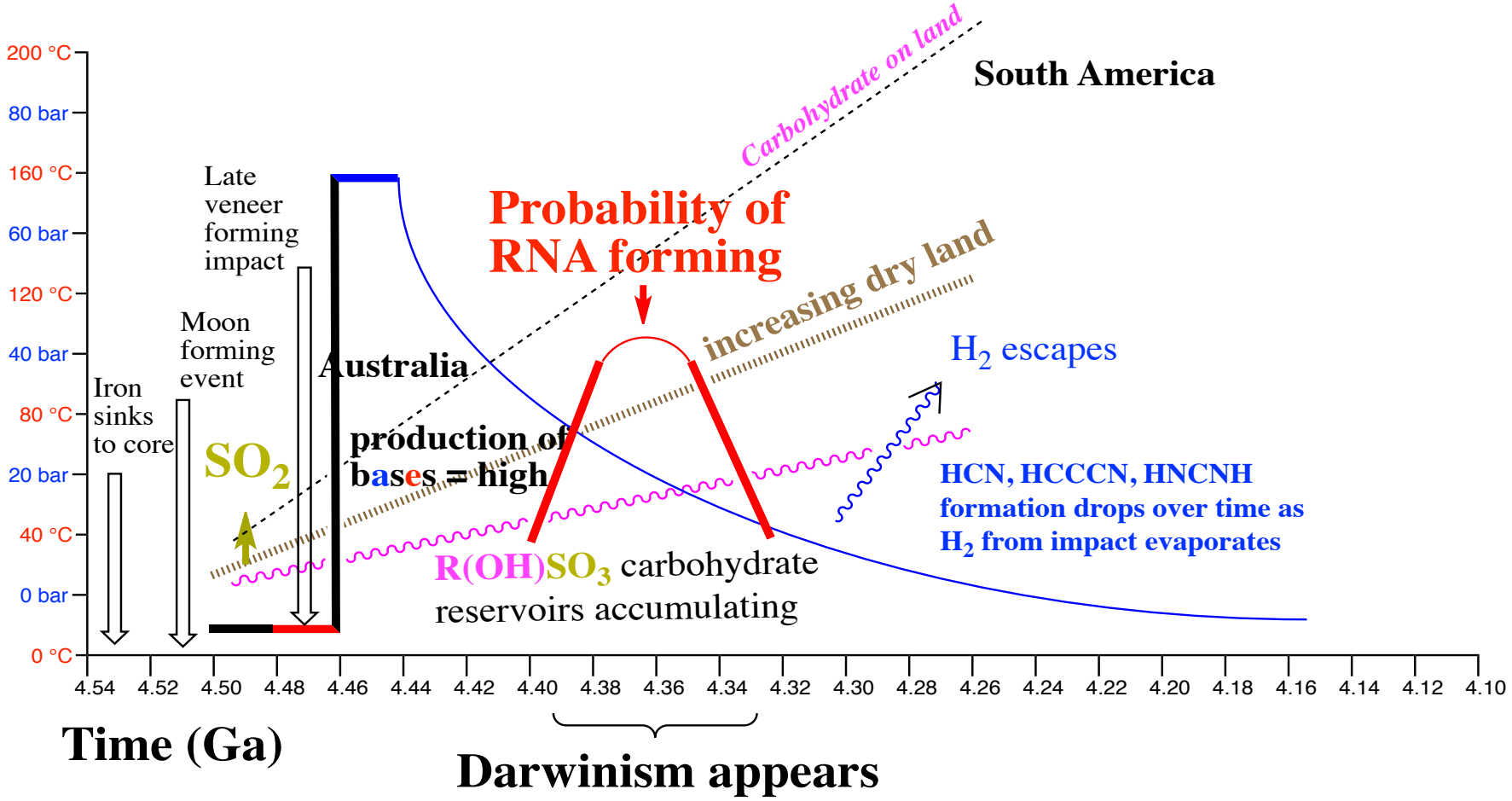
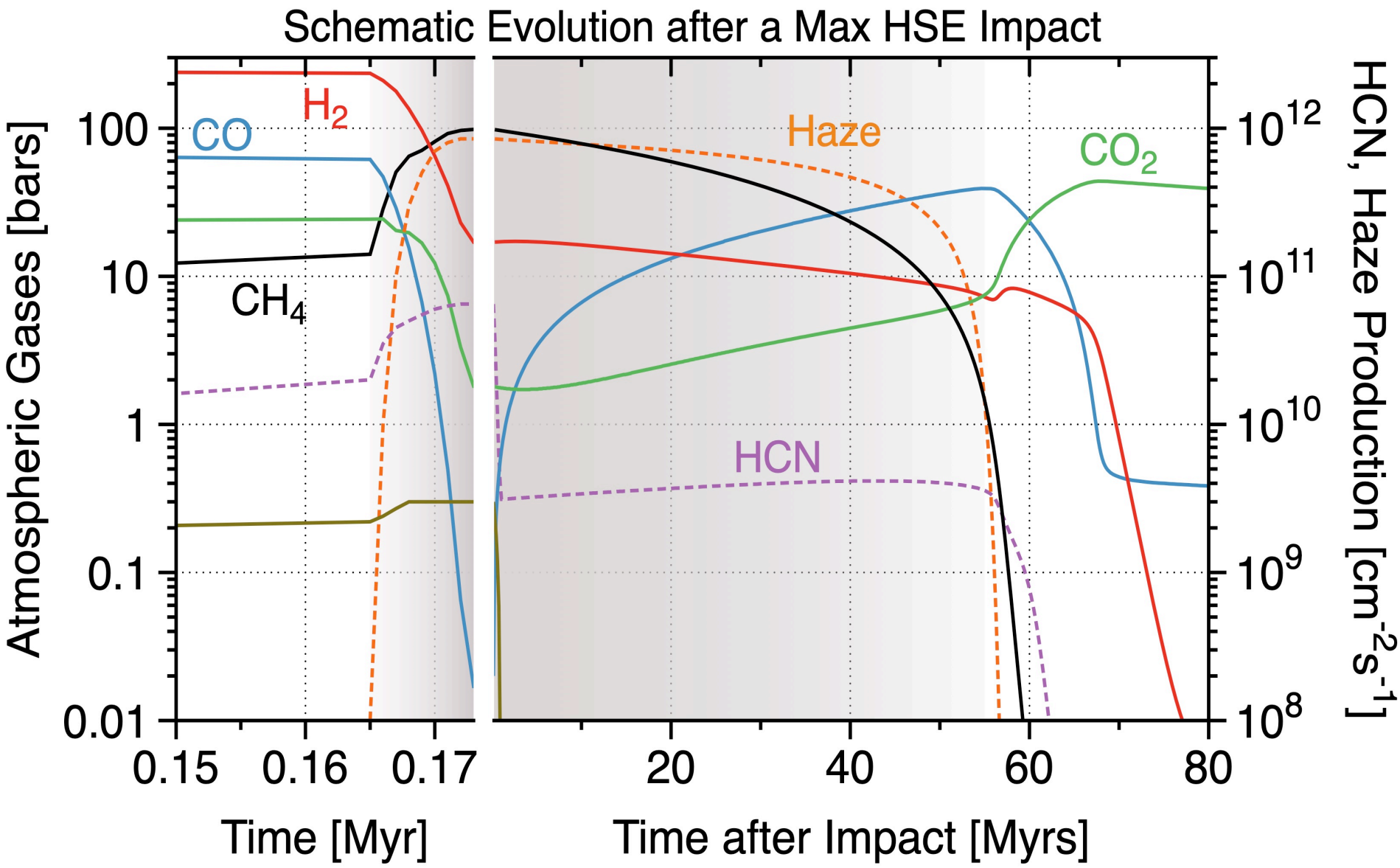


To understand the error, we must understand its source, here a rate of decay of a productive reducing atmosphere,

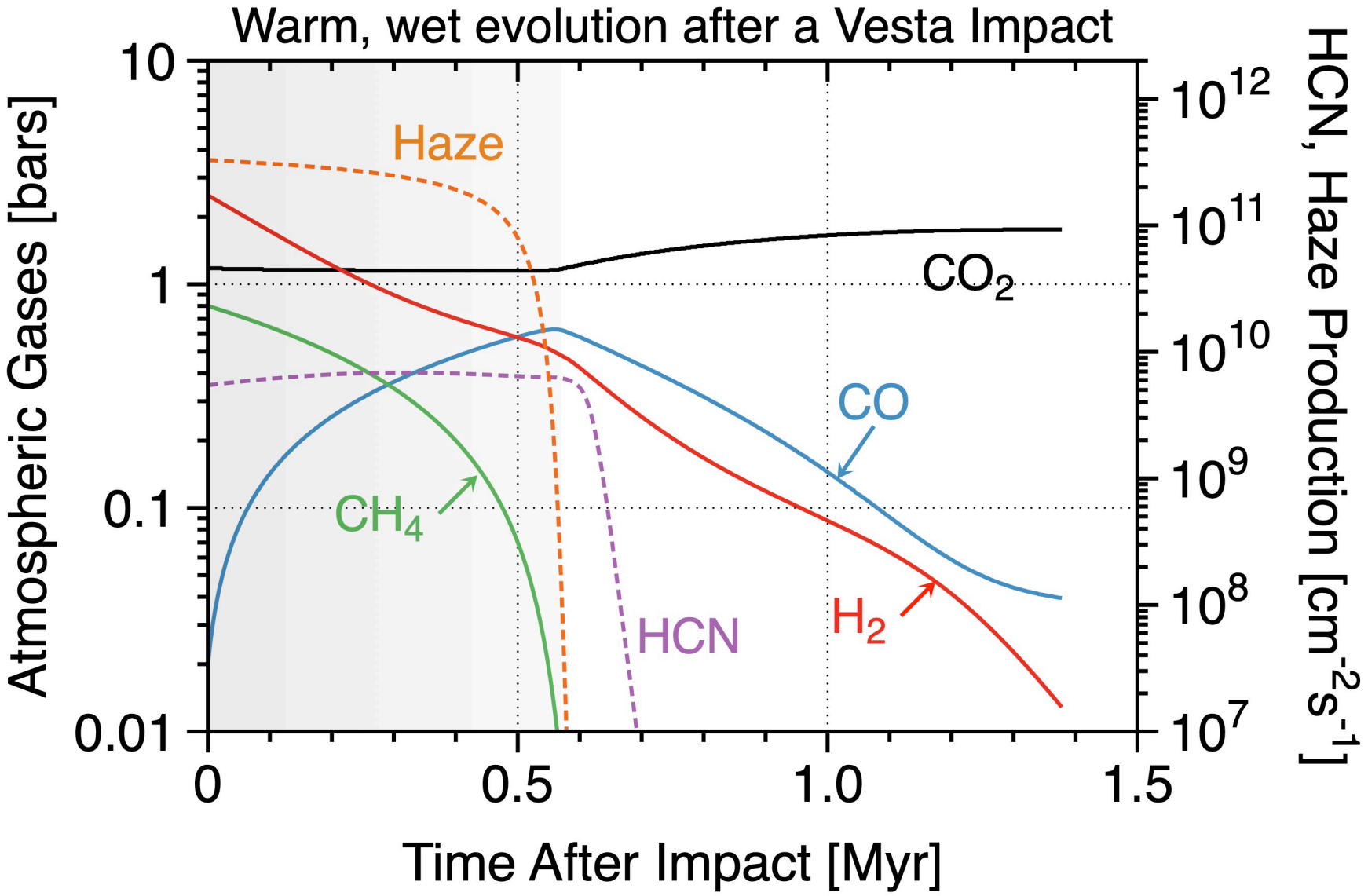
Rate of loss of productive atmosphere depends on many things (Solar intensity, various composition parameters) but time constant not 100 MY



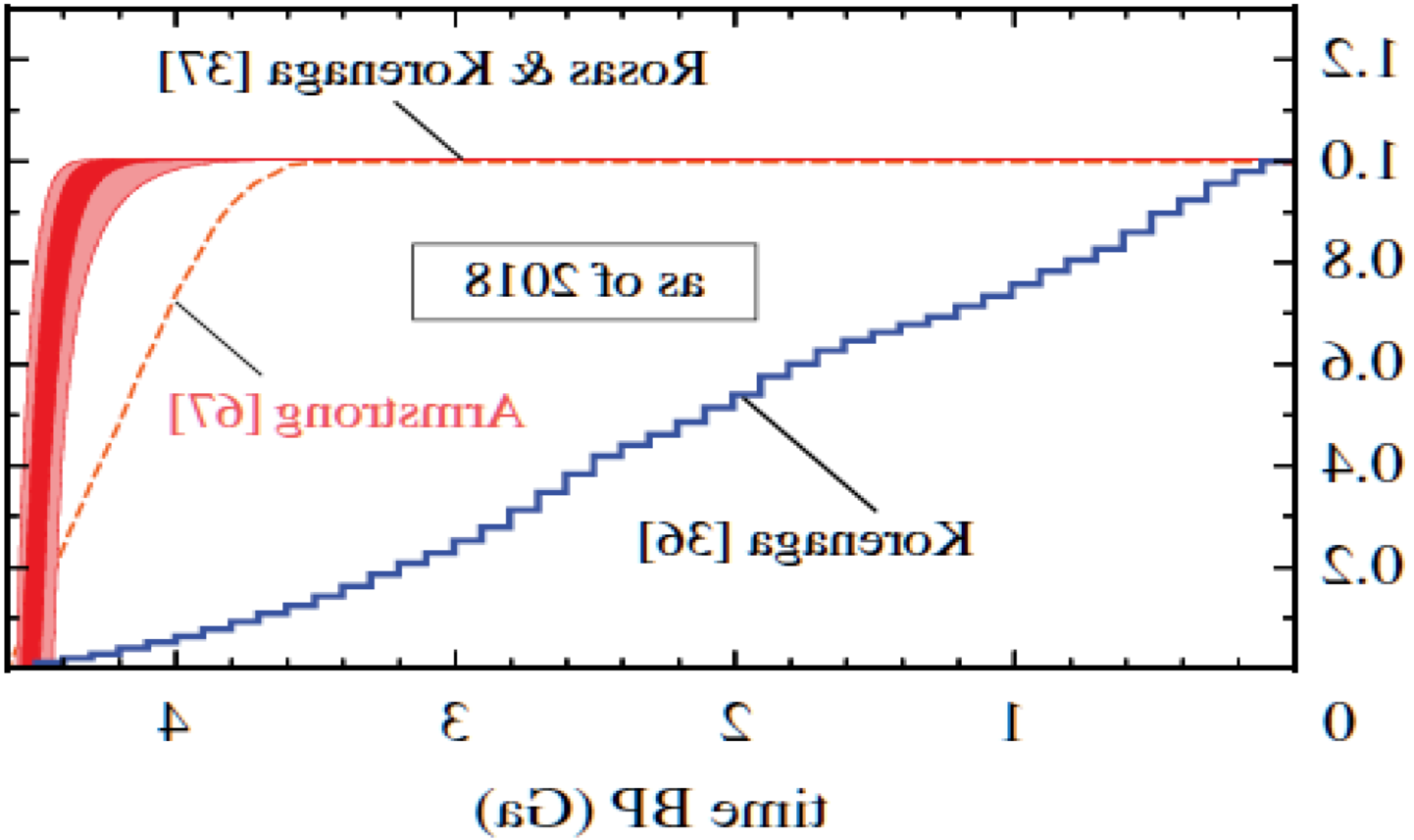


Time constant for decay for the Moon-size impactor is irrelevant if a later impactor did the sterilizing ...

Vesta is maybe smallest sterilizing impactor (Ceres better), but here the opportunity window for productive synthesis is ± 0.5 MY



In fact, the biggest source of uncertainty is the availability of dry land, as you learned yesterday from Jun Korenaga and Dustin Trail



Remembering, the amount of crust ≠ the amount of dry land.
Reducing the oceans to H_2 with iron will increase dry land.

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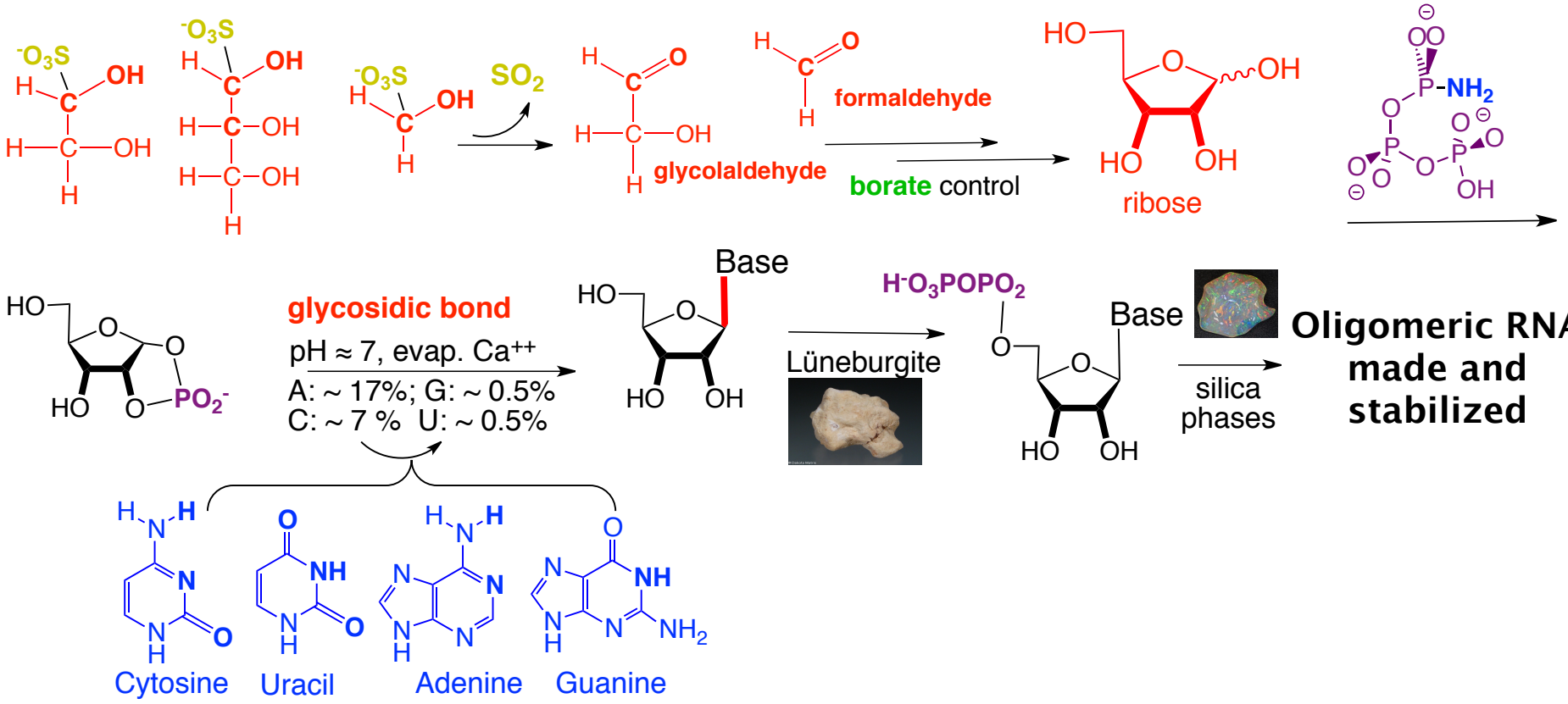
Next round of problems/paradoxes



The **borate** carbohydrate processing model is not complete enough

We need to know more about availability of Hadean **borate**

We have not said a word about chirality

A range of paradoxes arise from the poverty of RNA as a catalyst

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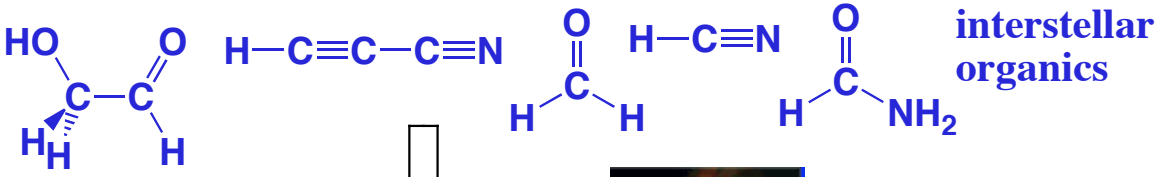
Understanding this thing called "life"

Geology added to organic chemistry

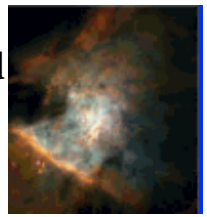
Prebiotic Chemistry



interstellar organics

Ricardo *et al*. (2004) *Science* **303**, 196

forward in time



Benner (2004) *Acc. Chem. Res*. **37**, 784-797

Info from natural history

A path to the simplest first life

understand better the possibilities

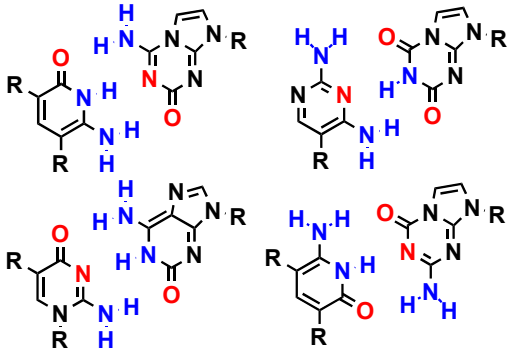
an independent genesis?

Explore Solar System

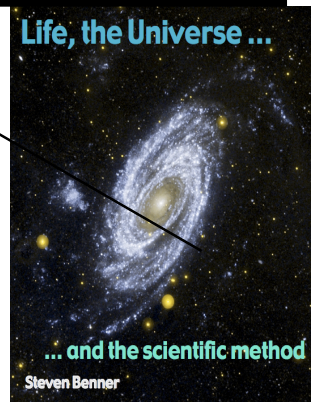
Baross, Benner, et al. (2007) *Limits of Organic Life in Planetary Systems*

First system to support Darwinian evolution

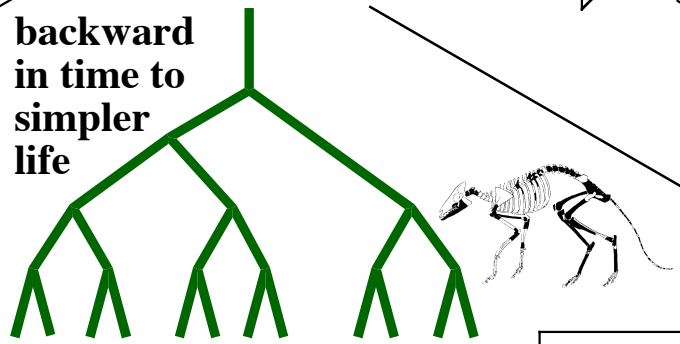
Construct alternatives in the lab



Synthetic biology



backward in time to simpler life



infer ancestral life forms; resurrect for laboratory study

Paleogenetics

Benner *et al*. (2007) *Adv. Enzymol. Mol. Biol. Protein Evol*. **75**, 1-132

Evolution ←→ mechanistic enzymology

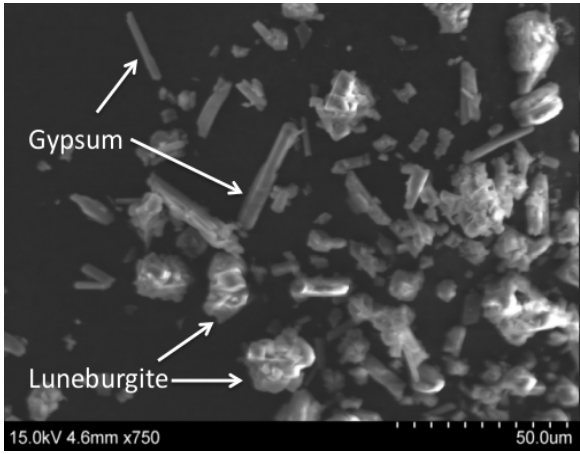
Calcium, sulfate, magnesium, borate, phosphate segregate to give lüneburgite

Ca^{+2} PO_4^{-3} ⇩ apatite

SO_4^{-2} Mg^{+2} ⇩ epsomite

Ca^{+2} SO_4^{-2} ⇒ gypsum

PO_4^{-3} BO_3^{-3} Mg^{+2} ⇒ lüneburgite



Lüneburgite plus nucleosides makes nucleoside diphosphates