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The CAS Conference on Molecular Origins of Life is the opening event of the CRC 235 "Emergence of Life" with Dieter Braun as spokesperson and also supported by the Simons Foundation.

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WELCOME ADDRESS

Dear participants,

A very warm welcome to the CAS Conference on "Molecular Origins of Life" 2018 in Munich. The conference takes place in the context of the CAS Research Group „Recreating the Origin of Life".

Munich has a long tradition in fostering interdisciplinary research on that topic. The focus of this conference is to bring together international and especially young scientists to join forces to experimentally approach the origins of life on Earth.

The complex origins of life on Earth can only be understood through the combined efforts of researchers in a wide range of disciplines in the natural sciences. The multifaceted nature of the problem is exemplified by the fact that experts from many different fields such as biophysics, astrophysics and chemistry are attending this international conference "Molecular Origins of Life", which is held now for the second time in Munich.

The conference will have short talks followed by discussions. We aim to open up the field to a broader audience, including the scientists who are not working in this particular field, but who are interested in the topic. The 24 talks in the program will cover topics such as how the first genetic material might have appeared, how one might rebuild a primordial cell, and what kinds of chemicals might serve as precursors for the evolution of living systems on other planets.

Due to the successful launch of the Collaborative Research Center (CRC) 235 "Emergence of Life", funded by the DFG for the next 4 years, we will be able to host this conference also in 2020 and 2022, hopefully starting a long term series.

We are looking forward to exciting discussions and to hearing about new experiments in various disciplines that could bring us a step closer to solving the mystery of how living systems originate. So please join the ride to understand the other discipline's standpoints on the origins of life and be inspired for a new round of ever better connected experiments.

The origins of life is a puzzle – let's try to connect the pieces!

Dieter Braun

PROGRAM

Thursday, 11 October 2018

Session A

08:30 - 8:55	**Job Boekhoven (TU Munich)** Self-selection of dissipative assemblies from primitive chemical reaction networks
8:55 - 9:20	**Irene Chen (UC Santa Barbara)** Fitness landscapes of RNA
9:20 - 9:45	**Dora Tang (MPI for Cellular Molecular Biology and Genetics)** Synthetic cellular models for probing the origin of life
9:45 - 10:05	**Discussion**
10:05 - 10:35	*Coffee break*

Session B

10:35 - 11:00	**Clemens Richert (University of Stuttgart)** Enzyme-free replication of genetic sequences
11:00 - 11:25	**Phil Holliger (MRC Laboratory of Molecular Biology)** Replicating RNA with RNA
11:25 - 11:50	**Niles Lehman (Portland State University)** Scrambling to build the RNA world
11:50 - 12:10	**Discussion**
12:10 - 13:30	*Lunch break*

Session C

13:30 - 13:55	**François Guyot (Museum National d'Histoire Naturelle, Paris)** Emergence of organized geochemical systems in the early earth

13:55 - 14:20	**Christof Mast (LMU Munich)** Physical non-equilibria as driving force and habitat for the origin of life
14:20 - 14:45	**André Estevez-Torres (ESPCI Paris & Sorbonne Université)** Synthesis of spatio-temporal structures with DNA molecular programs
14:45 - 15:05	**Discussion**
15:05 - 15:35	*Coffee break with* **poster session**
15:35 - 16:35	**Poster session**

Session D

16:35 - 17:00	**Andrei Lupas (MPI for Developmental Biology)** Ribosomal proteins as documents of the transition from unstructured polypeptides to folded proteins
17:00 - 17:25	**Sijbren Otto (University of Groningen)** Can we synthesize life in the lab? How chemistry may become biology
17:25 - 17:50	**Laurie Barge (California Institute of Technology)** Simulating prebiotic chemistry in hydrothermal systems on early earth and ocean worlds
17:50 - 18:10	**Discussion**
18:10	End

Friday, 12 October 2018

Session E

08:30 - 8:55	**Dimitar Sasselov (Harvard University)** Stellar UV light and the origins of life
8:55 - 9:20	**Cornelia Meinert (Université Nice Sophia Antipolis)** The asymmetry of life
9:20 - 9:45	**Stephen Mojzsis (University of Colorado)** Onset of late accretion to the inner solar system – consequences for the first habitats
9:45 - 10:05	**Discussion**
10:05 - 10:35	*Coffee break*

Session F

10:35 - 11:00	**Matt Powner (University College London)** On the divergent synthesis of purine and pyrimidine nucleosides
11:00 - 11:25	**Saidul Islam (University College London)** Prebiotic selection and assembly of proteinogenic peptides and ribonucleotides
11:25 - 11:50	**Thomas Carell (LMU Munich)** The prebiotic origin of RNA building blocks on early earth
11:50 - 12:10	**Discussion**
12:10 - 13:40	*Lunch break*

Session G

13:40 - 14:05	**Matthew Pasek (University of South Florida)** Disequilibrium polyphosphate formation from phosphorus redox
14:05 - 14:30	**Sheref Mansy (University of Trento)** The emergence of iron-sulfur catalysts
14:30 - 14:55	**Shawn McGlynn (ELSI of Tokyo Institute of Technology)** How much can we learn about ancient cells from sequence analysis?
14:55 - 15:15	**Discussion**
15:15 - 15:45	*Coffee break*

Session H

15:45 - 16:10	**Robert Pascal (University of Montpellier)** How chemical kinetics can become a driving force for life self-organization
16:10 - 16:35	**Moritz Kreysing (MPI of Molecular Cell Biology and Genetics)** Ribozyme reactions in the presence of uncharged and charged co-polymers
16:35 - 17:00	**Frank Postberg (Universtiy of Heidelberg)** The subsurface ocean of enceladus: a habitable place in our solar system
17:00 - 17:20	**Discussion**
17:20	Closing of the Conference

Coffee/tee/cold drinks and snacks are available during the coffee breaks in the atrium. Unfortunately no lunches and dinners can be offered. You will find many restaurants in walking distance.

TALKS

Thursday, 11 October 2018

Friday, 12 October 2018

Session A

Thursday, 11 October, 8:30

Self-selection of dissipative assemblies from primitive chemical reaction networks

Marta Tena-Solsona, Caren Wanzke, Benedikt Riess, Job Boekhoven
Department of Chemistry, Technische Universität München and
Institute for Advanced Study, Technische Universität München

Life is a dissipative non-equilibrium structure that requires constant consumption of energy to sustain itself. How such an unstable state could have been selected from an abiotic pool of molecules remains a mystery. [1] Recent work has demonstrated the emergence of life-like features in highly out-of-equilibrium non-biological systems. Features that include bistability and oscillations, [2] the formation of molecular assemblies [3] that can undergo dynamic instabilities [4] or can be controlled over space and time, [5] yet efficient selection mechanisms need to be identified.

Here, we show that phase separation is an efficient way for a controlled selection of non- equilibrium chemical species. We bring a library of primitive carboxylic acids out of out- of-equilibrium by condensing agents. We find the metastable anhydride products that can phase separate into micron-sized droplets to be more persistent against degradation than non-assembling ones. After several starvation-refueling cycles, the library had self-selected the most competitive product by their survival in phase separated droplets. The observed self-selection can be fully rationalized by a first order kinetic model, taking into account the persistence of species by compartmentalization. Our results suggest that sophisticated auto-catalytic behavior is not a necessity for the selection in non-equilibrium assemblies.

[1] R. Pascal, A. Pross, J. D. Sutherland, Open. Biol., 2013, 3, 130156
[2] S. Semenov, L Kraft, A. Ainla, M. Zhao, V. Campbell, K. Kang, G. Whitesides, Nature, 2016, 537, 656.
[3] S. van Rossum, M. Tena-Solsona, J. van Esch, R. Eelkema, J. Boekhoven, Chem. Soc. Rev., 2017, 46, 5519.
[4] J. Boekhoven, W. E. Hendriksen, G. J. Koper, R. Eelkema, J. H. van Esch, Science., 2015, 349, 1075
[5] M. Tena-Solsona, B. Rieß, R. K. Grötsch, F. C. Löhrer, C. Wanzke, B. Käsdorf, A. R. Bausch, P. Müller- Buschbaum, O. Lieleg, J. Boekhoven, Nat. Commun., 2017, 8, 15895

Session A

Thursday, 11 October, 8:55

Fitness Landscapes of RNA

Irene Chen
Department of Chemistry and Biochemistry, University of California, Santa Barbara

Evolutionary outcomes are difficult, if not impossible, to predict, largely because the effect of any possible mutation is unknown. In other words, understanding evolution requires detailed knowledge of the relationship between sequence and activity, or the fitness landscape. Inspired by the RNA World theory of early life, in which RNA would carry information and also perform catalytic functions, we study the emergence and evolution of functional RNAs. Our experimental efforts focus on mapping complete fitness landscapes [1] of ribozyme activity, as well as estimating the fitness distribution of random RNA [2]. We also study how encapsulation in a vesicle affects RNA activity and structure [3]. These studies inform our understanding of the likelihood of emergence of function and the roles of chance vs. natural selection in the evolution of the RNA World.

[1] Jimenez et al., Proc. Natl. Acad. Sci. USA 110:14984 (2013)
[2] Pressman et al., Nuc. Acids Res. 45:8167-8179 (2017)
[3] Saha et al., Nat. Commun. 9:2313 (2018)

Session A

Thursday, 11 October, 9:20

Synthetic cellular models for probing the origin of life

Dora Tang
Max Planck Institute for Cellular Molecular Biology and Genetics, Dresden

The onset of Darwinian evolution could have coincided with primitive compartmentalization which would have concentrated reactive molecules capable of genetic propogation or metabolism. However, the precise nature of the molecules which drive self-assembled compartmentalization are unknown. Molecular fingerprints from modern biology give some clues to the types of interactions which might have been important for primitive compartmentalization. To this end we use a multidisciplinary approach for the bottom-up synthesis of dynamic protocellular systems for origin of life studies.

In this talk I will describe the synthesis of artificial cells based on liquid-liquid phase separation (coacervation) and hydrophobic effects such as lipid vesicles and discuss the plausibility of their roles as protocellular models [1,2]. Specifically, I will describe how these coacervate microdroplets may be used to concentrate and localize molecules and support primitive reactions based on RNA [3].

[1] J. Vieregg et al., Curr Opin Colloid Interface Sci. 26, 50 (2016)
[2] T-Y Dora Tang et al. Nat Chem. 6(6), 527 (2014)
[3] Drobot et al. BioRxiv 273417; doi: https://doi.org/10.1101/273417 (2018)

Session B

Thursday, 11 October, 10:35

Enzyme-free replication of genetic sequences

Clemens Richert, Elena Hänle, Daniel Pfeffer, Marilyne Sosson, Sebastian Motsch, Peter Tremmel, Eric Kervio
Institute of Organic Chemistry, University of Stuttgart

Transmitting genetic information to daughter cells is critical for all known forms of life. To do so in the absence of enzymes is difficult because the reactivity of present-day nucleotide building blocks is too low to get significant copying yields and because the base fidelity can be low [1], possibly leading to an „error catastrophe“. Other issues that complicate enzyme-free replication scenarios, such as strand separation, have also been identified [2]. We have recently reported the first enzyme free replication system using chemically activated aminonucleotides and reversible termination [3]. This model system requires intervention by the experimental scientist employing it, adding fresh aqueous solutions at predefined time points, but it allows an unprecedented look at the fidelity of genetic replication with nucleotides in the absence of enzymes. Using mass spectrometric monitoring, we found that replication systems employing just G and C perform much better than those using all four canonical bases of DNA [3]. We are currently extending this work to experimental systems involving unmodified RNA and ribonucleotides that are activated in situ [4]. Issues such as regioselectivity of phosphodiester formation, extension to enzyme-free ligation, and the formation of peptido RNAs under genetic copying conditions [5] will also be discussed.

[1] E. Kervio, A. Hochgesand, U. E. Steiner, C. Richert, Proc. Natl. Acad. Sci. 2010, 107, 12074
[2] J. W. Szostak, J. Syst. Chem. 2012, 3, 1
[3] E. Hänle, C. Richert, Angew. Chem. Int. Ed. 2018, 57, 8911
[4] M. Jauker, H. Griesser, C. Richert, Angew. Chem. Int. Ed. 2015, 54, 14559
[5] H. Griesser, P. Tremmel, E. Kervio, C. Pfeffer, U.E. Steiner, C. Richert, Angew. Chem. Int. Ed. 2017, 56, 1219

Session B

Thursday, 11 October, 11:00

Replicating RNA with RNA

James Attwater, Edoardo Gianni, Chris Wan, Isaac Gallego, Philipp Holliger
MRC Laboratory of Molecular Biology, Cambridge

A critical event in the origin of life is thought to have been the emergence of an RNA molecule capable of self-replication as well as mutation, and hence evolution towards ever more efficient replication. Although this ancestral replicase appears to have been lost, key functional aspects of RNA-catalyzed RNA replication can be studied “by proxy” with the use of modern RNA enzymes (ribozymes) generated by in vitro selection.

Starting from the R18 RNA polymerase ribozyme, a descendant of the class I ligase ribozyme derived directly from a random RNA sequence pool, we have used both in vitro evolution and RNA engineering to generate new RNA polymerase ribozymes with improved polymerase activity and sequence generality. I’ll be presenting our progress in the engineering and evolution of RNA polymerase ribozymes towards a general polymerase and self-replication capacity.

We have discovered RNA polymerase ribozymes that are capable of the templated synthesis (i.e. transcription) of another simple ribozyme [1] or long RNA oligomers on a favourable template [2]. Recently, we also discovered that simple peptides rich in lysine (or its simpler analogues) are able to potentiate RNA polymerase ribozyme activity and evolution [3]. I’ll also be presenting our work on the potential role that structured media such as the eutectic phase of water ice [4] - as well as physicochemical cycles such as freeze-thaw cycles [5] - may have played in early RNA evolution and catalysis and the emergence of functional RNAs from the pools of short RNA oligomers accessible through prebiotic chemistry.

Finally, I’ll be presenting recent work on the engineering of triplet polymerase ribozymes that are able to copy and replicate even highly structured RNA templates and enable non-canonical reverse and primer-free replication modes [6].

[1] Wochner A et al., Science 332, 209 (2011)
[2] Attwater J et al., Nat Chem 5, 1011 (2013)
[3] Tagami S et al., Nat Chem. 9, 325 (2017)
[4] Attwater J et al., Nat Commun 1, 76 (2010)
[5] Mutschler H et al., Nat Chem 7, 502 (2015)
[6] Attwater J et al., eLife, 7:e35255 (2018)

Session B

Thursday, 11 October, 11:25

Scrambling to build the RNA world

Benedict Small, Bryce Clifton, Ryo Mizuuchi, Niles Lehman
Department of Chemistry, Portland State University

On a primordial Earth, during the origins of life, pre-life polymers would have had to undergo processes both to elongate their lengths and to enrich their sequence diversity. The RNA world hypothesis provides a testable framework for the study of how such polymers can originate, evolve, and develop into dynamical pre-biological systems. Recombination, the swapping (or scrambling) of large blocks of genetic information in an energy-neutral fashion, provides a plausible means for RNA populations to increase in length and diversity [1, 2, 3] in the absence of protein enzymes. Here, we investigate a variety of old [4] and new means by which spontaneous RNA-directed recombination of RNA can occur in vitro. Specifically, we have discovered two novel mechanisms of RNA recombination involving the attack of the 2′, 3′, and 5′ hydroxyl groups of one RNA strand on another to create new sequence combinations. Computer simulation of such events describes how novel and complex RNAs can arise in an ecological fashion from simple oligomers. We thus conclude that there are facile means by which RNA could have bootstrapped itself towards biological complexity.

[1] N. Lehman. Chem. Biodivers. 5, 1707–1717 (2008)
[2] N. Vaidya et al. Nature 491, 72–77 (2012)
[3] A. Blokhuis & D. Lacoste. J. Chem. Phys. 147, 094905 (2017)
[4] A. V. Lutay et al. Chem. Biodivers. 4, 762–767 (2007)

Session C

Thursday, 11 October, 13:30

Emergence of organized geochemical systems in the early earth

François Guyot
IMPMC, Muséum National d'Histoire Naturelle, Paris

Living cells are out-of-thermodynamic-equilibrium systems using mostly organic molecules bathed in ionic aqueous medium as a material support. In order to understand their emergence in the early Earth, one way is to examine geochemical environments in which (1) organic matter was available independently of any pre-existing life, (2) sources of disequilibrium were continuously available, and (3) a coupling mechanism existed between those disequilibria and organic chemical reactions. In this work, I will show a thermodynamic and kinetic model of abiotic organic matter evolution in geochemical environments of the early Earth based on analogs found in present day geological media. One result is that abiotic organic synthesis on Earth is difficult below 200°C although possibilities at lower temperatures are currently actively investigated. For the emergence of the molecular systems of life, temperature gradients in oceanic or continental hydrothermal settings (those latter being compatible with photochemistry) are particularly interesting since they provide simultaneously: (1) continuously sustained disequilibria on time scales of thousands of years, (2) efficient abiotic organic synthesis, and (3) good possibilities of concentration of diluted reactants [1]. Another result of this model is that iron-sulfur geochemistry provides a unique way of coupling temperature gradients with specific organic synthesis. I will use experimental results demonstrating that a related mechanism is indeed operating in hyperthermophilic archaea (Thermococcales) that populate hydrothermal vents [2]. Moreover, the pervasive presence of Fe3S4 greigite-related Fe-S clusters in several fundamental proteins of metabolism poses quite strong constraints on the natural media from which these general biological mechanisms might have emerged in relation with a thermodynamic modelling of greigite formation that will be explained.

These geochemical characteristics, however, point to a great difficulty: how to understand the ubiquity of phosphate chemistry in biomolecules (e.g. [3])? Some evidences indeed exist of decoupled histories of sulfur-based and phosphate-based chemical networks in present day living systems [4]. Thermodynamic and kinetic models for understanding the association of sulfur-based and phosphate-based systems will be discussed in the framework of a global prebiotic chemical reactor [5].

[1] P. Baaske et al., PNAS. 104, (2007)
[2] A. Gorlas et al., PloSOne, (2018)
[3] M.A. Pasek et al., Chem. Geol. 475. (2017)
[4] J.E. Goldford et al., Cell 168, (2017)
[5] E.E. Stüeken et al., Geobiology. 11, (2013)

Session C

Thursday, 11 October, 13:55

Physical non-equilibria as driving force and habitat for the origin of life

Christof Mast[1], Michael Kieß[1], Friederike Möller[1], Noel Yeh Martin[2], Corinna Kufner[3], Stefan Krebs[4], Mara L. Heinlein[1], Matthias Morasch[1], Hannes Mutschler[5], Helmut Blum[4], Wolfgang Zinth[6], Sheref Mansy[2], Dieter Braun[1]
[1] Systems Biophysics, LMU München,
[2] CIBIO, University of Trento,
[3] Harvard-Smithsonian Center for Astrophysics, Harvard University,
[4] Gene Center, LMU München,
[5] Max Planck Institute for Biochemistry, Martinsried,
[6] Biomolecular Optics, LMU München

Life can be abstracted as a non-equilibrium steady state, driven by externally generated energy fluxes. Four billion years ago it was likely that living processes were triggered by existing non-equilibria such as phonon/heat or photon fluxes. We are interested how such systems were initially able to influence random molecules of prebiotic chemistry in sequence, chirality and function. We found that thermal gradients across water filled pores lead to a concurrent fluid convection and directed movement of dissolved charged molecules along the temperature difference. Combined, both effects accumulate the dissolved biomolecules in a length dependent manner, which coagulates mutually interacting oligonucleotides. This process depends on the oligomer sequence and chirality: A mixture of strands with different sequence demixes into sequence-pure and homochiral coagulates upon thermal accumulation, implementing a possible selection pressure for favoring interacting strands during the origin of life.
The thermal non-equilibrium also creates and maintains a pH gradient over two units by the selective accumulation of charged buffer molecules, which shifts the local equilibrium in pH. In this system, early compartments of life may have cycled between different external pH conditions, implementing a continuous proton flux across their boundaries which could possibly be converted to energy rich chemicals.
Interestingly, thermal gradients over water-air interfaces also appear to have a positive effect on complex systems such as protein-expressing cell-free extracts. This could indicate a physical complementation function of gas bubbles, which at the same time provide a working space for complex biomolecule systems under a wider range of external conditions.
We also use high throughput sequencing to analyze UV induced dimeric lesions on DNA and RNA. In a streamlined process, damage rates for all possible dimer and trimer sequences were determined in agreement with literature. The results allow for a quick determination of future targets of chemical simulations and might reveal effects of early UV radiation on the genetic code.

[1] C.B. Mast et al., PNAS 110(20), 8030-5 (2013)
[2] C. B. Mast and D. Braun, PRL 104, 188102 (2010)
[3] M. Morasch et al., ANIE 55/23, 6676-6679 (2016)
[4] L. Keil et al., Nature Communication 8, 1897 (2017)

Session C

Thursday, 11 October, 14:20

Synthesis of spatio-temporal structures with DNA molecular programs

Jean-Christophe Galas[1], Anton Zadorin[1], Yannick Rondelez[2], Georg Urtel[1], Anis Senoussi[1], André Estevez-Torres[1]
[1] Laboratoire Jean Perrin, CNRS and Sorbonne University, Paris,
[2] Laboratoire Gulliver, CNRS and ESPCI, Paris

Biological systems combine two levels of molecular complexity. First, they synthesize molecular structures with exquisite chemical properties. Second, they construct out-of-equilibrium chemical reaction networks displaying capabilities that are uncommon in man-made molecular systems: measure time and space, and compute. Chemistry has a longstanding history of synthesizing complex molecular structures. The question that motivates our research is: what level of spatio-temporal complexity can we attain in a dissipative chemical system?

In an attempt to answer this question, I will describe a systems chemistry approach relying on the programmability of DNA. Firstly, I will introduce a set of highly reconfigurable synthetic chemical reaction networks based on DNA. Secondly, I will describe how we have used it to synthesize different spatio-temporal concentration patterns, such as travelling waves [1,2] and stationary fronts [3]. Finally, I will discuss the possibility of exploiting these patterns for fabricating materials inspired from embryonic development.

[1] Padirac A et al (2013) Spatial waves in synthetic biochemical networks. J. Am. Chem. Soc. 135(39):14586-14592
[2] Zadorin AS, et al (2015) Synthesis of programmable reaction-diffusion fronts using DNA catalyzers. Phys. Rev. Lett. 114(6):068301
[3] Zadorin AS, et al (2017) Synthesis and materialization of a reaction–diffusion french flag pattern. Nature Chemistry 9:990

Session D

Thursday, 11 October, 16:35

Ribosomal proteins as documents of the transition from unstructured polypeptides to folded proteins

Vikram Alva, Hongbo Zhu, Marcus D. Hartmann, Joana Pereira, Fabian Springer, Jörg Martin, Andrei N. Lupas
Department of Protein Evolution, Max Planck Institute for Developmental Biology, Tübingen

Folded proteins are the essential catalysts of life, but the ability to fold is a rare, complicated, and easily disrupted property, whose emergence at the origin of life is poorly understood. We have proposed that folded proteins arose from an ancestral set of peptides that acted as cofactors of RNA-mediated catalysis and replication [1]. Initially, these peptides were entirely dependent on the RNA scaffold for their structure, but as their complexity increased, they became able to form structures by excluding water through hydrophobic contacts, making them independent of the RNA scaffold. Their ability to fold was thus an emergent property of peptide-RNA coevolution.
The ribosome is the main survivor of this primordial RNA world. It's very slow rate of change makes it an excellent model system for retracing the steps that led to the folded proteins of today [2]. Towards its center, proteins are extended and largely devoid of secondary structure; further out, their secondary structure content increases and supersecondary topologies become common, although the proteins still largely lack a hydrophobic core; at the ribosomal periphery, supersecondary structures coalesce around hydrophobic cores, forming folds that resemble those seen in proteins of the cytosol. Collectively, ribosomal proteins chart a path of progressive emancipation from the RNA scaffold, offering a window onto the time when proteins were acquiring the ability to fold.

We retraced this emancipation for an αα-hairpin from ribosomal protein RPS20, which is unstructured in the absence of its cognate RNA, but which folds autonomously when repeated at least three times within the same polypeptide chain [3]. A global analysis of ribosomal proteins for fragments that could fold upon repetition or recombination shows that this is a wide-spread, albeit cryptic, property.

[1] V. Alva et al., eLife 4, e09410 (2015)
[2] A.N. Lupas & V. Alva. J Struct Biol 198, 74 (2017)
[3] H. Zhu et al., eLife 5, e16761 (2016)

Session D

Thursday, 11 October, 17:00

Can we synthesize life in the lab? How chemistry may become biology

Sijbren Otto
Centre for Systems Chemistry, Stratingh Institute, University of Groningen

How the immense complexity of living organisms has arisen is one of the most intriguing questions in contemporary science. We have started to explore experimentally how organization and function can emerge from complex molecular networks in aqueous solution [1]. We focus on networks of molecules that can interconvert, to give mixtures that can change their composition in response to external or internal stimuli. Molecular recognition between molecules in such mixtures leads to their mutual stabilization, which drives the synthesis of more of the privileged structures (Figure 1). As the assembly process drives the synthesis of the very molecules that assemble, the resulting materials can be considered to be self-synthesizing. Intriguingly, in this process the assembling molecules are replicating themselves, where replication is driven by self-recognition of these molecules in the dynamic network [2]. The selection rules that dictate which (if any) replicator will emerge from such networks are starting to become clear [3]. We have observed that factors such as mechanical energy [2] and the nature of the environment [4] can determine which replicator wins the competition for building blocks. We have also witnessed spontaneous differentiation (a process akin to speciation as it occurs in biology) in a system made from a mixture of two building blocks [5]. When such systems are operated under far-from-equilibrium flow conditions adaptation of the replicators to a changing environment can occur. Replicators that are able to catalyze reactions other than their own formation have also been obtained, representing a first step towards metabolism. Thus, the prospect of Darwinian evolution of purely synthetic molecules is tantalizingly close and the prospect of synthesizing life de-novo is becoming increasingly realistic.

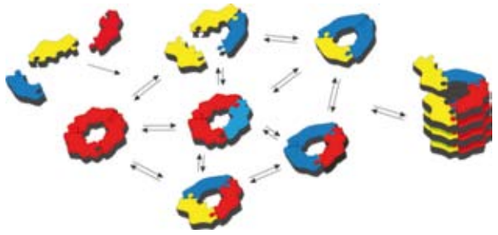

Figure 1 – Molecular recognition between molecules in a dynamic molecular network can lead to self-synthesizing materials, build up from self-replicating molecules.

[1] Li, J.; Nowak, P.; Otto, S. J. Am. Chem. Soc. 2013, 135, 25, 9222-9239
[2] Carnall, J. M. A.; Waudby, C. A.; Belenguer, A. M.; Stuart, M. C. A.; Peyralans, J. J.-P.; Otto, S. Science 2010, 327, 1502-1506
[3] Malakoutikhah, M.; Peyralans, J.J-P.; Colomb-Delsuc, M.; Fanlo-Virgos, H.; Stuart, M. C. A.; Otto,S. J. Am. Chem. Soc. 2013, 135, 49, 18406-18417
[4] Leonetti, G. Otto, S. J. Am. Chem. Soc. 2015, 137, 2067–2072
[5] J. W. Sadownik, E. Mattia, P. Nowak, S. Otto, Nature Chem. 2016, 8, 264–269

Session D

Thursday, 11 October, 17:25

Simulating prebiotic chemistry in hydrothermal systems on early earth and ocean worlds

Laurie M. Barge[1,2], Erika Flores[1,2], Keith Chin[1], John-Paul Jones[1], Ninos Hermis[1,2], Marc M. Baum[2,3]
[1] NASA Jet Propulsion Laboratory, California Institute of Technology, Pasadena,
[2] NASA Astrobiology Institute, Icy Worlds,
[3] Oak Crest Institute of Science, Monrovia

Serpentinization environments generate free energy driven by redox, pH, chemical, and thermal gradients; these gradients are known to sustain life in hydrothermal systems and may be significant for the emergence of life on Earth [1]. Similar hydrothermal systems may also be present on ocean worlds such as Europa and Enceladus, and therefore are of great interest to astrobiologists in the search for life [2,3]. Hydrothermal sediments and chimney precipitates in hydrothermal systems on the early Earth could have contained reactive minerals such as iron (-nickel) sulfides and iron oxyhydroxides. Behaving like flow-through chemical reactors, these mineral precipitates may have promoted various reactions towards the emergence of life including amino acid synthesis, concentration and retention of organic products, phosphorus redox and polymerization, and rudimentary energetic processes by electrochemistry. In vents today, and perhaps on the early Earth and / or other worlds, the precipitation of electrically active or reactive minerals can create geological fuel cell systems that may support life via electron transfer processes. We have utilized various experimental systems for simulating physicochemical gradients and geochemical redox reactions in serpentinizing systems, including the formation and characterization of simulated hydrothermal chimneys, synthesis of reactive hydrothermal sediments over a range of pH / redox states, and use of fuel cells as planetary habitability test-beds to simulate redox geochemistry of vents [4,5]. I will discuss these experimental approaches for simulating astrobiologically relevant chemistry in hydrothermal vents, as well as how understanding serpentinization is significant for the search for life on other worlds.

[1] MJ Russell et al. (2014) Astrobiology 14:308–343
[2] S Vance et al. (2007) Astrobiology 7:987–1005
[3] H-W Hsu et al. (2015) Nature 519:207–210
[4] LM Barge et al. (2015) J Vis Exp 105, doi:10.3791/53015
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Session E

Friday, 12 October, 8:30

Stellar UV light and the origins of life

Dimitar Sasselov
Harvard University

The talk will discuss recent results on the astrophysical and planetary environmental context that makes possible the synthesis of precursors to RNA, peptides, and lipids. Our focus will be on the role of mid-range UV light from about 200 to 300 nm as a source of energy and as a very specific selection agent in chemical evolution.

Session E

Friday, 12 October, 8:55

The asymmetry of life

Cornelia Meinert
CNRS, Institute of Chemistry of Nice, University Nice Sophia Antipolis

What is responsible for the emergence of life's homochiral biopolymers – DNA/RNA and proteins – where all the constituent monomers exhibit the same handedness?

Based on in-situ observations and laboratory studies, we propose that this handedness occurs when chiral biomolecules are synthesized asymmetrically through interaction with circularly polarized light (cpl) in interstellar space [1]. Due to a true-chiral influence of cpl [2], the interaction with racemic or achiral molecules generates enantiomerically enriched mixtures. Our previous experimental results on the asymmetric photolysis of amino acids [3], as well as their absolute asymmetric synthesis from achiral interstellar ice precursor molecules [4,5], revealed polarization- and energy-controlled induced enantiomeric enrichments.

Furthermore, my recent research has shown that the central chiral unit of RNA, ribose, forms readily under simulated comet conditions (Fig.1) and this has provided new insights into the accessibility of precursors of genetic material in interstellar environments [6]. The significance of my research arises due to the current lack of experimental demonstration that amino acids and sugars can simultaneously and asymmetrically be synthesized by a universal physical selection process. In my presentation, I will therefore highlight a few significant results on our on-going cometary ice simulation experiments, the chiroptical properties of targeted sugar and amino acid molecules in the UV using circularly polarized synchrotron light and present future strategies towards furthering understanding the origin of asymmetric prebiotic molecules.

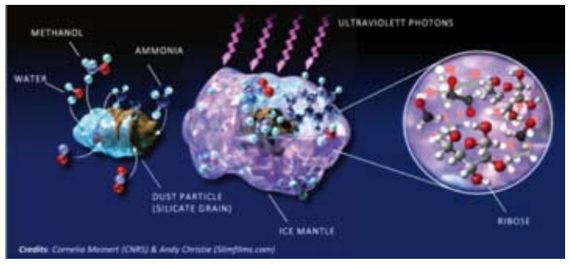

Figure 1 – Ribose forms in the icy mantles of interstellar dust grains from simple precursor molecules (water, methanol, and ammonia) under high energy radiation.

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Session E

Friday, 12 October, 9:20

Onset of late accretion to the inner solar system – consequences for the first habitats

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[2] Institute for Geological and Geochemical Research, Research Center for Astronomy and Earth Sciences, Hungarian Academy of Sciences, Budapest,
[3] Earth-Life Science Institute, Tokyo Institute of Technology,
[4] Planetary Science Institute, Tucson,
[5] Centre for Earth Evolution and Dynamics (CEED), University of Oslo, Collaborative for Research in Origins (CRiO), The John Templeton Foundation-FfAME Origins program

The timescale for late accretion to the terrestrial planets is poorly known. Nectarian, Tolstojan and Noachian (pre-3900 Ma) terranes are the oldest and most heavily cratered on the Moon, Mercury and Mars [1,2]. The latter two have crater densities vs. time comparable to the ancient highlands of the Moon as established from direct sampling in the Apollo and Luna missions. The earliest thermal events recorded in asteroidal meteorites show evidence for crust formation and subsequent continuous resetting by impacts of high closure-temperature radiogenic systems such as U-Pb and Pb-Pb up to about 4.43 Gyr [3]. This cooling age profile for asteroids pre-dates absolute ages for the last time the crusts of Earth (ca. 4.40 Gyr), Moon (ca. 4.42 Gyr) and Mars (ca. 4.43 Gyr), as established by U-Pb and (U-Th)/He zircon geochronology, could have experienced wholesale melting by bombardment [4,5,6]. Younger ages defined by the relatively low closure-temperature 40-39Ar geochronological system likewise display a continuum of ages, but these only pick up at ca. 4.48 Gyr and then extend to later times up to as young as 0.25 Gyr ago. Here, we couple dynamical models of late accretion, with ages compiled from radiogenic systems with variable sensitivity to age-resetting by thermal metamorphism, to show that subsequent to Moon formation the flux of comets to the inner solar system reset ages of planetary crusts to ca. 4.45 Ga. Concurrent bombardment by leftover planetesimals continued to impact the inner solar system following a smooth (monotonic) decline in flux afterwards. We describe the dynamical basis of this phenomenon in the context of giant planet migration and assess the likelihood that biospheres could have been established – and continue to survive – beginning about 150 Myr after solar system formation on Earth and Mars.

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[3] D.D. Bogard Chem. Erde 71, 207 (2011)
[4] R. Brasser et al., Astrophys. J. 821, 75 (2016)
[5] R. Brasser et al., Earth Planet Sci. Lett. 455, 85 (2016)
[7] N.L. Kelly et al., Earth Planet Sci. Lett 482, 222 (2018)

Session F

Friday, 12 October, 10:35

On the divergent synthesis of purine and pyrimidine nucleosides

Matthew W. Powner
Department of Chemistry, University College London

RNA is the leading candidate for the first biopolymer of life, due to its dual biological role in information transfer and catalysis, as well as the deep-seated evolutionary history of non-coding RNA (e.g. 16S and 23S ribosomal genes, tRNA genes, nucleotide binding domains).
Although remarkable progress has been made toward understanding prebiotic nucleotide synthesis, to date all syntheses have accounted separately for pyrimidine and purine ribonucleotides [1,2]. Two divergent syntheses, which furnish pyrimidine and purine nucleosides from a common prebiotic precursor, are presented [3,4,5,6]. A generational and constitutional relationship between 2,2′-anhydropyrimidines and 8,2′-anhydropurines is proposed to enable the divergent and concomitant synthesis of nucleosides [5]. First, the divergent prebiotic synthesis of pyrimidine and 8-oxo-purine ribonucleotides, from one reaction sequence, with regiospecific glycosidation and complete furanosyl selectivity is presented [5]. Then, a selective hydrogen sulfide-mediated purine reduction that provides a mechanism to furnish the complete set of Watson-Crick arabinonucleosides is described [6]. Photochemical purine reduction is observed to be highly efficient for the desired canonical purines (A, G), but highly destructive for the non-canonical inosine, suggesting UV-light may have played a role in nucleobase selection en route to extant life. The unity of the decribed reaction pathways indicates that consolidation of these two strategies, which furnish RNA's canonical sugar moiety and RNA's canonical bases, respectively, holds the key to elucidating the origins of ribonucleotides in biology.

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Session F

Friday, 12 October, 11:00

Prebiotic selection and assembly of proteinogenic peptides and ribonucleotides

Saidul Islam
Department of Chemistry, University College London

The biological relationship of peptides and nucleotides is a strong indication that they have a common prebiotic origin [1,2], but their selective synthesis require high-purity monomeric building blocks. The intractable mixtures resulting from indiscriminate and uncontrolled chemical reactivity en route to these structures is a long-standing problem, which has thwarted the formation of the canonical peptide and RNA monomers [2]. Multistep pathways require the sequential addition of reactants and purification of intermediates that are consistent with reasonable geochemical scenarios. Ideally, building blocks for proteinogenic peptides and canonical nucleotides would be specifically selected, even from complex mixtures. Selective crystallisation offers one of the simplest and most plausible physiochemical means of prebiotic purification [3].
This talk will describe how a simple, inconspicuous, sulfur-bearing heterocycle, which is a likely by-product of mixed nucleic- and amino acid syntheses, resolves several long-standing problems caused by complex prebiotic mixtures [4]. 2-Aminothiazole acts as a chemical 'chaperone', guiding selective synthesis of peptide and nucleotide precursors from highly complex mixtures, and sets the stage for the emergence of canonical nucleotides [5, 6] and proteinogenic peptides in a completely traceless manner. Finally, the chemoselective prebiotic ligation of peptides in water will be described [7]. Collectively, these results augment a growing body of evidence that points towards the powerful and emerging role of sulfur in prebiotic chemistry, and reinforces the notion that peptides and nucleotides share a unified chemical origin.

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[7] Unpublished

Session F

Friday, 12 October, 11:25

The prebiotic origin of RNA building blocks on early earth

Sidney Becker, Thomas Carell
Department of Chemistry, LMU München

The widely accepted RNA world hypothesis suggests that life first emerged from RNA, which is able to (self)-replicate and evolve. Replication of RNA requires formation of the complementary pyrimidine-purine Watson-Crick base pairs A:U and G:C, which are a prerequisite for accurate genetic information transfer. Although prebiotic pathways to RNA building blocks have been reported, no pathway has been able to generate all four constituents of RNA simultaneously.[1, 2] We recently discovered a new pathway (FaPy-pathway) that is able to generate purine nucleosides under plausible prebiotic conditions.[3] The formation of the purine RNA building blocks is driven exclusively by fluctuations of physicochemical parameters such as pH, temperature and concentration. These conditions allowed in addition the parallel formation of a variety of non-canonical purine nucleosides as living molecular fossil of an early abiotic world.[4] Many of the formed non-canonical RNA building blocks are today assumed to have been part of the genetic system of the last universal common ancestor (LUCA).[5] We therefore suggest that these non-canonical nucleosides were formed in the abiotic phase of the origin of life. In order to find a prebiotically plausible scenario for the parallel formation of purine and pyrimidine bases to create the fundamental Watson-Crick base pairing system, we developed new prebiotically plausible chemistry to pyrimidines that are compatible with our purine procedures. This was meant to provide all four RNA building blocks in the same geochemical environment.[6] We found that the reaction of cyanoacetylene with hydroxylamine is a perfect starting point. It creates first amino-isoxazoles in high yields from which the pyrimidines are easily formed. The new chemistry also affords the naturally occurring 5′-mono- and 5′-di-phosphorylated ribosides exclusively as their furanosides. This observation now provides a plausible explanation for why these thermodynamically disfavoured furanosidic constitutional isomers are exclusively present in the backbone of RNA and DNA. Our results show that all central constituents of RNA could have been part of the same prebiotic nucleoside/tide pool, as a prerequisite for RNA to evolve on early Earth. Our chemistry suggest that the formation of RNA on the early earth was not guided by chance but an inevitable consequence of early earth chemistry.

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Session G

Friday, 12 October, 13:40

Disequilibrium polyphosphate formation from phosphorus redox

Matthew A. Pasek
School of Geosciences, University of South Florida, Tampa

The chemistry of phosphorus includes the biochemistry of replication and metabolism. In these reactions, phosphate esters and acid anhydrides control specific aspects of the chemistry of major molecules, from their solubility to reactivity to their stereochemistry. The prebiotic origin of such molecules is unclear, and indeed such molecules may not even have been prebiotic [1]. Here, I propose a new route to production of polyphosphates as phosphorylating agents. This proposed reaction involves the reduction of phosphate (P5+) to phosphite (P3+) by a concomitant oxidation of iron (Fe2+ to Fe3+) followed by the preferential extraction of phosphite by water [2]. Phosphite is stable to oxidation under most conditions, until it interacts with a highly oxidizing radical such as OH [3]. Under such conditions, phosphite may react to form metaphosphate (PO3-), which may in turn react with phosphate to form pyrophosphate, and subsequently larger polymers such as polyphosphate and cyclic trimetaphosphate. In such an environment, phosphorylation of organics leading to nucleotides may be feasible. I report here the newest results on the oxidation and phosphorylation of organics by these polyphosphates, suggesting that the early earth may have been predisposed to a prebiotic, phosphorylating environment [4].

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Session G

Friday, 12 October, 14:05

The emergence of iron-sulfur catalysts

Claudia Bonfio, Simone Scintilla, Noel Yeh Martin, Daniele Rossetto, Shibaji Basak, Sheref S. Mansy
CIBIO, University of Trento

Cells persist by balancing favorable chemistry with the thermodynamically unfavorable reactions that are needed to support the cell. In extant organisms, this balancing act is mediated in large part by iron-sulfur protein enzymes that work together with large proton pumps. However, the complexity of extant metabolic systems masks the chemistry that emerges from simple, prebiotically plausible mixtures of iron, sulfide, and short peptides. Together these ingredients spontaneously form iron-sulfur peptides under conditions compatible with the surface of the early Earth. The type of iron-sulfur cluster formed reflects the environmental conditions and suggests mechanisms for the templated formation of longer metallopeptides. The iron-sulfur peptides are catalytically active, capable of the iterative transfer of electrons leading to the formation of a pH gradient across membranes of the same magnitude found in living cells. As proton gradients are completely conserved across all known living systems, iron-sulfur peptide chemistry may have played an early role in shaping the chemistry that became metabolism.

Session G

Friday, 12 October, 14:30

How much can we learn about ancient cells from sequence analysis?

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[3] Max Planck Institute for Mathematics in the Sciences, Leipzig

A longstanding goal is to apply molecular phylogenetics to understanding ancient physiological and evolutionary states [1]. With the current explosion of molecular sequencing data [2], it is a good time to consider how far back we can peer with the comparative molecular lens, and ask if we can understand life in its nascent years. A standard test for inferring ancient genes is to analyze phylogenetic branch positions and determine if a gene separates the archaea and the bacteria, but many phylogenetic trees are cluttered by phenomena such as horizontal gene transfer and non-orthologous displacement [3], making inferences difficult. In this presentation, we will give an overview of recent work in this area [4, 5], and also present new analyses which aim at relating inter-domain transfer (IDGT) to alignment sequence entropy. We find that sequence plasticity viewed through the lens of information entropy varies between functional groups of proteins and also within groups. Moreover, IDGT can be viewed in relationship to sequence alignment entropy, revealing how information changes (or doesn't) in response to IDGT. These findings will be discussed in the context of inferring the characteristics of the most ancient cells.

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Session H

Friday, 12 October, 15:45

How chemical kinetics can become a driving force for life self-organization

Robert Pascal
IBMM, CNRS, University of Montpellier, ENSCM, Montpellier

The emergence and developments of life associated with the self-organization of increasingly improbable and complex systems challenge the usual view that the 2nd Law rules the evolution of material systems. Though it is widely accepted that open systems held far from equilibrium by receiving an input of low entropy energy can evolve towards a local decrease in entropy without violation of the 2nd Law [1], this understanding does not provide a driving force towards self-organization that remains better understood through the Darwin's theory of evolution. In a purely chemical approach to the origin of life, Addy Pross introduced the concept of Dynamic Kinetic Stability (DKS) [2] to account for the driving force for the evolution of chemical (and biological) systems capable of reproducing themselves toward higher degrees of complexity through the selection of variants. The kinetic requirements for the search for improved DKS to become the determining factor of the evolution of the system will be analyzed as well as their consequences on the establishment of favorable conditions for the origin of life.

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[3] R. Pascal and A. Pross, Chem. Commun. 51, 16160 (2015)
[4] R. Pascal et al., Open Biol. 3, 130156 (2013)
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[6] R. Pascal, Isr. J. Chem. 55, 865 (2015)

Session H

Friday, 12 October, 16:10

Ribozyme reactions in the presence of uncharged and charged co-polymers

Mrityunjoy Kar[1,2], Juan Manuel Iglesias Artola[1,2], Björn Drobot[1], Stefan Tassoulas[1,2], Kristian Le Vay[3], Oliver Beutel[1], Alf Honigmann[1], Dora Y. Tang[1], Hannes Mutschler[3], Moritz Kreysing[1,2]
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Ribozymes are usually studied at high concentration and at high purity.
Diluting ribozymes with their substrates below their dissociation constants strongly reduces their activity, which is frequently the case already in the high nanomolar regime. Rescuing activity by increasing affinity has the drawback of irreversible substrate binging, which leads to lasting ribozyme inhibition. As an alternative, we show that chemically inert co-polymers, e.g. PEG of various lengths, can rescue ribozyme activity in the dilute regime.
Specifically, we find that both for ligases and cleaving ribozymes, i) the initial velocity of the reaction is increased, ii) reactions last longer, and iii) they typically result in higher yields. Activity enhancement is robust again salt variations. We further find that, reaction rescue is threshold-length dependent, but as we show, it is neither due to surface passivation of reaction vessels by PEG, nor due to precipitation. Instead we suggest that ppm level PEG acts as a co-factor in ribozyme folding into their active confirmations. This suggestion is substantiated by FRET measurements.
Furthermore, we asked the question if the rescue effect could also be achieved with peptides. Here we observe that soluble, charged peptides lead to complex formation and ultimately coacervation, with a strong tendency to inhibit ribozyme reactions. Currently, we investigate how charge density mediates ribozyme mobility in order to recover activity.

Session H

Friday, 12 October, 16:35

The subsurface ocean of enceladus: a habitable place in our solar system

Frank Postberg[1,2], Nozair Khawaja[1,2], Fabian Klenner[1,2], Lenz Nölle[1,2], Ralf Srama[3], Hsiang-Wen Hsu[4], Jack Hunter Waite[5], Gabriel Tobie[6]
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[2] Institute of Earth Science, Universität Heidelberg,
[3] Institute of Space Systems, Universität Stuttgart,
[4] Laboratory of Atmospheric and Space Physics, University of Colorado, Boulder,
[5] Southwest Research Institute, San Antonio,
[6] Laboratoire de Planétologie et Géodynamique, University of Nantes

Saturn's icy moon Enceladus harbours a global ocean [1], which lies under an ice crust and above a rocky core [2]. Through warm cracks in the crust a cryo-volcanic plume ejects ice grains and vapour into space providing access to materials originating from the ocean [3,4]. Hydrothermal activity is suspected to be occurring deep inside the water-percolated porous core [5,6] and powered by tidal dissipation [7]. Two mass spectrometers aboard the Cassini spacecraft, the Cosmic Dust Analyzer (CDA) and the Ion and Neutral Gas Spectrometer (INMS) frequently carried out compositional in situ measurements of plume material emerging from the subsurface of Enceladus.
Our latest results now show that some emitted ice grains contain concentrated macromolecular organic material with molecular masses clearly above 200u [8]. The mass spectra of the two instruments provide key constraints on the macromolecular structure that contains both aromatic and aliphatic constituents as well as oxygen bearing and likely nitrogen bearing functional groups. The finding is suggestive of thin organic-rich film on top of the oceanic water table [8].
We suggest that the detected organics originate from Enceladus' hydrothermally active rocky core. Thermal ocean convection together with bubbles of volatile gases, transports these and other materials from the moon's core up to the ocean surface. There, organic nucleation cores - generated by bubble bursting and subsequently coated with ice from vapor freezing - are ejected into space. This nucleation shows similarities to the formation of ice clouds from organic sea spray on Earth and allows probing of Enceladus' organic inventory in drastically enhanced concentrations.

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POSTERS

Thursday, 11 October, 15:05 – 16:35

P1 Investigation of RNA degradation under early earth conditions

Amine Akermi[1,2], Barbara M. Giuliano[2], Paola Caselli[2], Hannes Mutschler[3], Birgitta Müller[2]
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Can simple genetic systems based on polynucleotides such as RNA survive under conditions provided by early earth or even space? Are these environments even able to support primitive biocatalysis and evolution? To address these questions experimentally, we combine our expertise in astrochemistry, molecular spectroscopy, and RNA (bio-)chemistry. Using a powerful multi-functional reaction chamber capable of simulating a wide range of environmental conditions, we have established a versatile in situ methodology allowing us to observe RNA building blocks with high molecular resolution during simulations using infrared (IR) spectroscopy.
Here, we present a first pilot study, in which we monitor the effect of UV irradiation on the IR-signatures of short RNA homopolymers (A12 and U12) under aqueous and frozen (-10°C) conditions. We observe that the IR-signatures of A12/U12 mixtures show weaker resistance to photodamage compared to isolated strands, changing more rapidly over time under constant UV-irradiation compared to U12 and A12. Moreover, freezing the samples above the eutectic point of water ice, which still enables RNA formation and catalysis, increases the overall apparent photostability of RNA.
In the future we will expand our research and combine it with powerful ex situ approaches to characterize RNA survival, synthesis, catalysis and evolution under plausible temperature, radiation, and chemical conditions present on early Earth and extra-terrestrial environments.

P2 Non-equilibrium phenomena in reaction-diffusion-advection systems

Bernhard Altaner, Patrick Hillenbrand, Ulrich Gerland
Fakultät für Physik, Technische Universität München

We study energy transduction processes in an out of equilibrium chemical reactor, i.e. the conversion between different forms of free energy that can occur in a dissipative reaction-transport system. In an autonomous well-mixed chemical reactor, a non-equilibrium steady state (NESS) can be maintained by chemostatting (i.e. buffering) the concentration of at least two species.
In contrast, we find that in a spatially extended environment a single buffered species coupling to a conservative force field is sufficient to keep the system out of equilibrium, if the buffered species participates in reactions. In that case the dynamic species form dissipative structures we call transport-reaction-cycles (TRCs), which are driven by spatio-chemical free energy differences.
In this work, we review some of the possible effects and thermodynamically characterize different forms of energy transduction in TRCs. For instance, we show how currents of charge carriers can be used to drive transport currents and enhance chemical reactions like polymerization. Further, certain kinetic limits are discussed analytically.

P3 How ancient is sense-antisense coding?

Zachary Ardern, Stefan Wichmann, Alina Glaub, Siegfried Scherer
Department of Microbiology, Technische Universität München

The standard genetic code theoretically enables more than one protein to be encoded within the same nucleotide sequence in different reading frames. For instance, the sequence encoding a protein could be embedded within another protein-coding gene, in one of the three possible antisense frames. The general phenomenon of ‚overlapping genes' is well attested in viruses [1,2], but examples have also been discovered in cellular organisms [3,4]. The hypothesis that many ancestral proteins at the very beginning of protein coding were encoded in antisense to each other has been discussed in the literature with particular reference to amino acyl tRNA synthetases [5], but has not gained widespread acceptance. Usually the hypothetical primordial sense-antisense coding and observed modern day overlapping genes are not discussed together, but whether they could be connected should be investigated. Is sense-antisense encoding of proteins a trait that was found in the last universal common ancestor and perhaps earlier, or is it restricted to a few relatively recently derived tips of the tree of life? To begin to answer this question, we assess the taxonomic distribution and ages of antisense overlapping gene pairs, implement a recently developed algorithm [6] to test whether ancient protein domains are better able to encode proteins in antisense, and explore the relationship between sense-antisense coding and the structure of the standard genetic code.

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P4 Catalytic activity of prebiotic metallo-peptides

Shibaji Basak, Sheref Mansy
CIBIO, University of Trento

Enzymes are complex, large protein molecules that successfully perform the catalytic transformation of simple sugar molecules to high energy phosphate molecules in metabolism. These high energy phosphates are used to convert ADP to ATP, which is the main source of energy in cells. It was found that some peptides have catalytic activity and could represent prebiotic peptide analogues, potentially displaying modern day-like activity. Therefore, small peptides could have played a significant role in the evolution of life. We are engaged in finding small metallo-peptides, in particular iron-sulphur peptides that have the ability to catalytically oxidize metabolites of catabolic pathways. Short peptides have been identified that can act on glycolytic intermediates and funnel electrons down thermodynamically favourable paths. We seek to connect this protocatabolic reaction to electron transfer events that give rise to a pH gradient across lipid membranes.

P5 Prebiotic origin of all four RNA building blocks

Sidney Becker, Hidenori Okamura, Stefan Wiedemann, Thomas Carell
Department of Chemistry, LMU München

The widely accepted RNA world hypothesis suggests that life first emerged from (self)-replicating RNA. Replication requires formation the complementary pyrimidine-purine Watson-Crick base pairs A:U and G:C, which are a prerequisite for accurate genetic information transfer. Although separate prebiotic pathways to either purine or pyrimidine RNA building blocks have been reported, no pathway has been able to generate all four constituents of RNA simultaneously.[1, 2] Therefore, the spontaneous evolution of a Watson-Crick base pairing system is highly problematic. We report a prebiotically plausible pathway that provides all four RNA building blocks in the same geochemical environment.[3] The chemistry also affords the naturally occurring 5′-mono- and 5′-di-phosphorylated ribofuranosides with high selectivity. This provides a plausible explanation for why these thermodynamically disfavoured constitutional isomers are exclusively present in the backbone of RNA and DNA. Our results show that all central constituents of RNA could have been part of the same prebiotic nucleoside/tide pool, as a prerequisite for RNA to evolve on early Earth.

[1] Becker et al., Science 352, 833 (2016)
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[3] Becker et al. (submitted)

P6 Investigation of a class of prebiotic plausible organocatalysts for photochemical alkylations

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Selectively modifying simple, prebiotically plausible molecules, e.g derived from Fischer-Tropsch-Synthesis, is a powerful tool to extent available molecular structures. In this project we introduce a new class of organocatalysts, formed under prebiotic conditions, which can be implemented in enamine, iminium and SOMO catalysis. The Imidazolidin-4-thiones evolve from the reaction of aldehydes or ketones with aminonitriles in the presence of hydrogen sulfide [1]. We built up a library of a diversity of structural variants and determined the selectivity of their formation by capillary electrophoresis. These catalysts were successfully applied in alkylation reactions of linear aldehydes by photo-induced organocatalysis, using solar radiation as the primary energy source on the Early Earth [2,3]. Due to the elongation and branching of aldehydes in the course of the catalysis, the latter can be employed for the synthesis of the second generation catalysts. The catalyst activity is compared among one generation, as well as during the evolution. Kinetic investigation of this complex reaction network is performed by NMR analysis.

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P7 Tracing primordial metabolism reflected by microorganisms under serpentinite-hosted systems via quantitative DNA stable isotope probing

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Serpentinite-hosted systems have been shown to be modern analogs for early Earth environments where life could have originated and evolved due to the presence of abiotic organic synthesis in the presence of strongly reducing and high pH fluids emanating from the serpentinization process of ultramafic rocks [1]. One of the key features of these harsh environments are the presence of deeply branching chemoautotrophic microbes, i.e. methanogens and acetogens, that are capable of fixing carbon using presumably ancient carbon fixation pathways [2,3] – a complete or partial reductive tricarboxylic acid (rTCA) and a reductive acetyl-CoA pathway. Here, we aimed to substantiate one of the hydrothermal origin of life hypotheses [2,3] via quantitative DNA stable isotope probing (qSIP) after amending 13C-labeled substrates such as bicarbonate, acetate and formate to the collected serpentinites and carbonates to better understand the active metabolism of existing, but underexplored microbes in potentially active hydrothermal environment where the methane anomalies were detected in the water column (St. Paul's Archipelago, Mid Atlantic Ridge – Expedition AL 170602). After a 24-hour incubation with 13CO2 and H2, the qSIP experiment showed that hydrogen stimulated carbon fixation by several uncultured chemoautotrophic microbes whose physiology remains poorly understood. This shows that the St. Pauls Rocks is possibly an active serpentinization system, capable of supporting life via H2 production from low-temperature and hydration of ultramafic rocks. Future plans are to investigate rTCA and reductive acetyl-CoA pathway functioning in additional serpentinization sites: Lost-City hydrothermal vent at the Mid-Atlantic Ridge [4] and the Costa Rica ophiolite dome [5].

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P8 Non-enzymatic cross-replication of DNA templates with in situ activated DNA oligonucleotides

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Possible mechanisms for non-enzymatic replication of oligonucleotides under prebiotic conditions are currently under an intensive investigation due to their importance early in the origin of life. Here we present a mechanism, whereby two DNA templates of 24 bases cross-catalyze each other's synthesis from four 12 bases long oligonucleotide substrates, two of which are in situ activated using a condensing agent EDC. Temperature cycling is employed to overcome product inhibition. Original DNA templates successfully amplify under serial dilution conditions achieving 2.4 fold amplification in 5 hours despite an EDC-induced inhibition via accumulation of catalytically inactive substrate. Detailed kinetics simulation of the system with experimentally measured rates closely explains the experimental data. It predicts that a rapid exponential amplification of DNA templates can be achieved when a recycling mechanism for EDC-modified substrates is found. Due to the formation of essentially natural phosphodiester bonds at the ligation sites, next-generation sequencing can be used in the next step to study the evolution of DNA information content from a random pool of substrates.

P9 Assembly of RNA/DNA origami hybrids and of small DNA tubes

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Assembly of nanostructures out of DNA and RNA strands plays a major role in nanotechnology as well as in fundamental research pertaining biochemistry and evolutionary biophysics. Considering the RNA world hypothesis [1] and the general potential of DNA self-assembly [2], two separate experimental setups were designed to better understand formation and possible functions of small structures made out of RNA and DNA strands.
In a first experimental setup, folding and stability properties of RNA-DNA hybrid structures were investigated by folding an mRNA scaffold strand (1102 nt) encoding eGFP using DNA oligonucleotide staples via the DNA origami technique. The scaffold was folded into a cylindrical six helix-bundle (6HB) with a length of 49 nm and a diameter of 7.2 nm.
In another experimental setup, assembly properties of small structures designed to mimic ribosomal catalytic effects in peptide bond formation were investigated. Short DNA tubes sized 9.4 and 12.9 nm in length and 7.3 nm in diameter were designed based on self-assembly of short oligonucleotides. The structure included a short RNA strand with a 5' phosphate group as part of one tube, and a binding pocket for reactions between amino acids and the 5' phosphate of this strand [3].
Future projects will involve investigations regarding possible catalytic effects of RNA/DNA-based structures that might have formed in a pre-ribosomal world. In a first step, a basic model based on DNA origami structures capable of binding short tRNA-mimicking DNA strands was designed. Apart from shedding light onto fundamental questions regarding the origin of life, the constructed setups may also open novel possibilities in other fields such as gene delivery.

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P10 Separation of complex monosaccharide mixtures and its application to the analysis of a formose network catalyzed by corroded Schreibersite

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Complex monosaccharide mixtures for instance arising from formose-type reactions are of great interest in the context of the Origin of Life and in meteorite and asteroid samples [1,2]. The separation and analysis of such mixtures is highly challenging due to the structural similarity and number of possible stereoisomers.

In a comparative study, we focused on several single- and two-step derivatization approaches for liquid and gas chromatographic separations. We developed a comprehensive derivatization protocol for monosaccharides based on a library of reference compounds ranging from formaldehyde to hexoses as well as reaction by-products or decomposition products such as polyalcohols and sugar acids. The combination of different methods allows the detailed analysis of carbohydrate composition in regard to substance class and enables identification of certain monosaccharides independent of the sample matrices [3].

Applying these methods, we have found the meteoritic material Schreibersite (Fe3P) to catalyze aldol reactions building higher monosaccharides from C1-C3 building blocks [4]. Schreibersite basifies the water in which it is immersed which together with released iron cations catalyzes reactions in the formose reaction network offering a prebiotic route to biologically relevant monosaccharides.

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P11 Formation of biomolecules under volcanic hydrothermal vent conditions and investigation with high-resolving analytical tools

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The Hadean earth formed about 4.8 billion years ago representing a hot planet with a lot of volcanic activities, regular comet bombardment, lack of oxygen and lack of life. This scenario does not look very friendly in our eyes but first forms of life were found in sediments about 3.5 billion of years old in western Greenland and Australia. Before we can find life in cellular form a certain set of biomolecules is needed. As a scenario for a chemical evolution, we choose volcanic hydrothermal vents, which offer a continuous supply of reactive nutrients and catalytic surfaces of transition metal minerals. The combination of high pressure and temperature leads to a high diversity of organic molecules including amino acids, hydroxy acids, fatty acids and small peptides [1-5]. High-resolving analytical tools and data-analytical approaches help to decipher these complex reaction mixtures [6-8]. The combination of Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR-MS), gas chromatography-MS (GC-MS) and nuclear magnetic resonance (NMR) methods have provided a complementary profile of the products for both low and high molecular weight compounds, including an enormous variety of CHOS derivatives. In summary, high-resolving analytical methods expand our view into the manifold chemistry of a potential origin of life scenario under volcanic hydrothermal vent conditions.

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P12 The effects of low pressure on DNA and lipid vesicles

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Studies on the size distribution of ancient fossilized raindrop imprints and on the isotope composition of fluid trapped in hydrothermal quartz indicate that during the late Archean eon (~3 billion years ago) the atmospheric pressure was lower than the modern levels with a value of ~0.5 bar as an upper limit [1,2]. At that pressure, the corresponding boiling temperature of water is lower, consequently affecting the temperature reachable by water surfaces. Moreover, evaporation and condensation rates, wet-dry cycles and drying processes are enhanced [3]. This means that many temperature dependent reactions (for example, heat induced DNA denaturation), have to be retaken into consideration in this low pressure scenario.
Here, we study the effects of a low pressure environment on macromolecules such as DNA, and lipid vesicles. In our experiments, we put our solution of fluorescently labeled molecules inside a 3D printed millimeter-sized chamber that is placed in a thermal gradient [4]. We make use of a vacuum pump to lower the pressure (down to ~0.2 bar), and directly observe this setting with a fluorescence microscope.
We find that a low pressure environment is able to trigger the separation of the two complementary strands of a 55bp DNA, as well as disaggregate lipid vesicles of oleic acid or DOPC. We propose that surface tensions at the air-bubble interface, shear flows induced by the boiling, low salt concentration in condensing droplets, and drying processes are drastic out-of-equilibrium processes that can trigger physical processes such as DNA denaturation and vesicle disaggregation in a low temperature regime.

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P13 RNA-peptide coacervates sustain ribozyme activity

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The RNA world hypothesis proposes that RNA molecules could have been used in a prebiotic scenario as enzymes and information storing molecules1. This hypothesis, however, lacks an explanation for the formation of compartments and has been traditionally studied under pure RNA conditions. Compartments can be formed using lipids to generate a membrane-barrier2 or through liquid-liquid phase separation (LLPS)3. For the latter case, complex coacervation, a kind of liquid-liquid phase separation, has two main advantages: (1) it can highly concentrate RNA molecules and (2) it generates a compartment that is readily able to exchange molecules with its surrounding. On the other hand, recent experiments hint that the interaction of RNA with peptides might lead to an increase in ribozyme activity4. Herein we show that a small ribozyme, the R3C ligase5, can form coacervates when combined with short lysine and arginine peptides. Furthermore, these coacervates are able to sustain enzymatic activity under certain conditions. We find that the material properties of the droplets depend on the strength of the peptide-RNA interactions and that these can be tuned by changing the peptide identity, size, charge density, and salt concentrations. When RNA-peptide interactions are strong, the coacervate phase shows gel-like properties, which arrest ribozyme activity. As interaction strength decreases, the phase becomes more liquid-like with enhanced RNA diffusion which rescues RNA activity. It is these more fluid coacervates the ones that sustain RNA enzymatic activity. We conclude by emphasizing the compelling role of peptide-RNA coacervates as a link between the RNA world and modern protocells.

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P14 Photochemical properties of oxazoline prebiotic precursors of RNA nucleotides

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Prebiotically plausible synthesis of RNA nucleotides and nucleosides has been one of the greatest challenges for the origins of life field [1]. At present, prebiotical reaction sequences are sought to demonstrate high-efficiency chemical routes leading to RNA monomers from credible feedstock and under the prebiotic conditions on the early Earth. In recent years, there have been many efforts to discover plausible reaction routes leading to the formation of RNA monomers. However, the most intuitive formation of N-glycosidic bonds between nucleobases and ribose proved to be highly inefficient for purines and practically impossible for pyrimidines. In addition, no selective and high-yielding sources of pure ribose were identified so far. To bypass the direct glycosilation, oxazolines having preformed N-glycosidic bond were proposed as the key intermediates and were successfully used to furnish ribonucleotides in the final parts of the syntheses [2, 3]. One of the crucial factors determining lifespan of molecules on the Archean Earth is photostability. Since UV-irradiation is an important element of many such prebiotically credible reaction sequences, the assessment of photochemical stability of oxazolines should be performed.
To scrutinize the photostability of arabino- aminooxazoline (aAO) and oxazolidinone thione (aOT), we performed computational explorations of excited-state potential energy surfaces to show mechanistic picture of non-adiabatic phenomena. Algebraic diagrammatic construction to the second order [ADC(2)] and complete active space perturbation theory (CASPT2) methods were used to carry out these calculations. Our results indicate efficient radiationless deactivation mechanisms of aOT and in conjunction with findings of UV-irradiation experiments, we anticipate that the listed precursor should be photostable on the Archean Earth. By contrast, our calculations suggest that aAO precursor does not absorb in the range of the sunlight spectrum that reached the surface of the early Earth. This indicates that aAO would survive in the prebiotic environment.

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P15 On the nature of earth's earliest known life forms on

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The theory of chemical evolution proposes that the self-assembly of organic compounds into protocells was a key event in the evolution of life. However, the question which has not been addressed is whether the existence of such proto-cells could actually have left behind a fossil trail? Here, we compare the morphologies of Precambrian microfossils with that of extant bacteria, after transforming them into liposome-like cells by manipulating the salt content of the media. In such a state, bacteria we tested seemed to have lost control over their morphology and cell division. Using this top down approach we were able to reproduce the morphologies of most known microfossils found in 3.7 - 1.8 Ga iron formations, many of which were considered to be morphologically too complex to be bacteria. Our work suggests that some of the most-studied Precambrian microfossils, so far annotated either as cyanobacteria or eukaryotic algae, most likely were liposome like proto-cells.

P16 Non-canonical enhancement of ribozyme activity by highly diluted (ppm) molecular crowder; polyethylene glycol

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The 'RNA world hypothesis' remains a hallmark in "origin of life" research despite very poor robustness and low reactivity of most model replicators studied so far. (1) Moreover, the 'RNA world', might have suffered from the debated 'dilution problem' before protocell formations. A frequently used trick to enhance ribozyme activity is the use of high concentration molecular crowders (M to mM) to increase RNA concentrations by excluded volume effects. (2) Here we show, that excluded volume effect is not strictly required to enhance ribozyme activity using the R3C ligase and Hammerhead as a model ribozyme and polyethylene glycol (PEG) as a model crowding agent.(3, 4) Polyethylene glycol is widely used as a crowder with the understanding that it has no interaction with ribozymes except by the excluded volume effect.(2, 4-6) Our results show that with ribozyme dilution, ppm levels of PEG significantly increase ribozyme activity. The Michaelis-Menten parameter (Km) of diluted reaction is significantly decreased in presence of 5 ppm PEG 10kDa. PEG is able to interact or change the molecular environment at low concentrations the ribozymes, which favors substantially (11-folds increase) the overall yield of the ligations. According to our results, impurities of 'inert' polymers could have had a functional role in pushing forward the ribozyme reactions in dilution.

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P17 Mapping the formose reaction network

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The base catalyzed self-condensation of formaldehyde, the so called formose reaction (FR), is the most accepted origin for the formation of carbohydrates on the early earth [1,2]. Since its discovery by Alexander Butlerow in 1861, the full distribution of intermediates and products as well as the mechanism of the reaction is still unknown and heavily discussed in literature [3,4]. Scrambling experiments with deuterated and undeuterated substrates in the FR gives us the opportunity to elucidate the reaction pathways in the entire reaction network. Separation of the products via GC followed by MSMS measurements gives unambiguous information about every step. Variation of the experimental settings like varying the temperature, the reaction time or the initial concentrations enables us to conduct kinetic studies. These results do not only give us a deeper understanding of this complex FR-network but also allow us to predict the conditions and parameters necessary for certain species to be formed.

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P18 Time resolved investigation of complex prebiotic mixtures by LC-MS/MS analysis

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Previous works showed the formation of ribonucleosides from simple carbon and nitrogen containing molecules [1,2]. Starting from 2-aminooxazole and glyceraldehyde, a network is formed which subsequently leads to the formation of cytidine and uracil. The product formation was herby proven via NMR-studies. Accomplishing the full elucidation of the given reaction network, liquid chromatography analysis techniques followed by downstream high-resolution QTOF mass analysis are applied to analyze the given reaction mixtures. Kinetic resolution of the single steps enables the determination of all formed products. Further, alteration of the reaction parameters gives additional information about the network.
In a next step, the role of the respectively used sugars prior to other formed sugars in the formose-reaction mixture is examined. This opens up deeper understanding of the reaction pathways finally leading towards ribonucleosides.

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P19 Sequence selection of oligonucleotides under a ligation chain reaction

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The replication of information on RNA or DNA is central for the emergence of life [1]. Previously, the replication of one sequence has often been in the focus, but we think it is essential to monitor the replication and selection dynamics of a broader pool of sequences and how it behaves under simple replication conditions.
We focus on the transition from template-free polymerization to templated ligation and consider the possibility, that the same chemistry for polymerization also triggered the first replication cycles by templated ligation. Once polymerization could create oligomers long enough to hybridize at a given temperature, we expect a nonlinear ligation dynamic to set in. We study whether sequences were selected at this onset of replication and if interesting non-linear and frequency-dependent behavior can be found [2].
For short strands, the ligation is dominated by the weak hybridization dynamics (ssDNA linked by Watson-Crick-base-pairing [3]).
By using AT-only (A: adenine, T: thymine) 12mer random sequences as starting material, the sequence space for the first ligation stage that creates 24mer can still be completely sampled. We found elongation to 36mers, 48mers and longer and could obtain more than 12 million individual strands using Next Generation Sequencing (NGS), showing a significant selection of sequences undergoing this elongation dynamics. We analyze the sequences with self-written LabView code and show how spiking with defined sequences changes the sequence selection dynamics of the replicated sequence pool.

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P20 Prebiotic sugar formation under mechanochemical conditions

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For the origin of life several building blocks like amino acids, nucleobases, carbohydrates and phosphates are necessary. The theory of the "RNA world" considers the RNA as universal molecule which contains for example ribose as sugar.[1] A prebiotic important route for sugar molecules is the formose reaction, a base-catalyzed condensation reaction. This reaction type represents several chemical and biologically relevant transformations, which are thermo¬dynamically disfavored in aqueous solution. In contrast, the proposed geochemical settings for the origin of life are, for instance, warm little ponds[2] and hydrothermal vents.[3]
As alternative, solvent-free approaches like mechanochemical reactions offer nonaqueous conditions. Prebiotic sources for mechano¬chemical energy can be lithospheric activity or meteoritic impacts on earth. For the realization in bench-scale ball mills are predestinated for the investigation of mechanochemical conditions.
We report on the formation of carbohydrates under nonaqueous conditions starting from glycolaldehyde and glyceraldehyde as substrates with a catalytic base present.[4] This part of the formose reaction network was investigated with and without mechano¬chemical energy input by use of a mixer ball mill. The complex product mixture was analyzed via gas chromato¬graphy-mass spectrometry.[5]

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P21 Microfluidic rock-like reactors to study the synthesis of the first nucleotides

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Amongst the many open questions in the origin of life is the origin of the first biomolecules: from lipids to amino acids to nucleotides, life exhibits a wealth of complex molecules whose pre-biotic synthetic pathways remain unclear. Whatever the answers, early biomolecules must have arisen under realistic geological boundary conditions. Recent chemical advances have shone light upon potential synthetic pathways for the production of nucleotides in the laboratory, starting from simple abiotic precursors [1]. However, and in spite of the latest progress [2], most experiments have been performed in bulk chemistry, and through independent and successive synthetic steps. Therefore, geological plausibility remains largely unexplored.
We have emulated the conditions of microfluidic flow in rock pores, to uninterruptedly drive, within one single system, the sequential reactions necessary to produce an activated nucleotide [1]. Using CAD software and finite-element-method simulations, we have designed microfluidic devices to run the nucleotide synthesis autonomously, and to recreate a scenario analogous to what could be found in porous rocks on early Earth. We produce the reactors assembled from a repertoire of modular building blocks using stereolithographic 3 D printers, allowing for many applications beyond our own. The flow can be driven by gravity, increasing the geological realism of our system. Our methods also allow us to embed rock powders into the microfluidic system, enabling the testing of potential pre-enzymatic catalysts and further increasing geological plausibility. These results open the door to studies of any scenarios for the emergence of life, whereby chemistry is driven in a geologically realistic system to generate the earliest biologically relevant molecules.

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P22 Hypothesis, proposal and some estimates for enhanced abiogenesis experiment

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Based on the works of Prigogine and von Hayek hypothesis was proposed which allowed developing a new scheme for the origin of life experiments - the logical improvement of classic Miller's experiment in the form of „micro-planet" installation [1]. The improvement is aimed to production of wider variety of gradients, degrees of freedom and hence possible ways of development for prebiotic chemical processes. The scheme provides cycles of wetting-drying, evaporation-condensation-solidification etc. in a small confined space along with providing the two-phase mixing and presence of fresh and „ocean" water in conjunction with a variety of minerals, foam etc. It also supposes excitation of the system by different kinds of energy fluxes with various time periods. Different compositions and/or cycling programs can be compared by periodically monitoring of the laser light absorption and scattering spectra [2] richness, as well as by in vitro nanoparticles size measurement with correlation spectroscopy [3]. The installation basic parameters and components, as well as the possibilities of its application, are discussed.

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P23 Genetic exchange between protocells

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Early protocells are thought to have consisted of a self-replicating, RNA-based genetic system and a primitive metabolism enclosed within an amphiphilic membrane [1]. These protocells would have relied on simple self-organization processes presumably driven by environmental events. One such process, likely critical for early cellular evolution, is the exchange of content such as genetic material between individual protocells.
Using DNA-containing giant unilamellar vesicles (GUVs) as model systems, we investigated freeze-thaw cycles as an environmental driver for content exchange between vesicles [2]. We tested various freezing and thawing scenarios and, under certain conditions, found content mixing between vesicles to be very thorough with evidence thereof in all observed vesicles. Surprisingly, we found that content is transferred through diffusion across the membranes of tightly packed GUVs, and not through vesicle fusion and fission, as was observed in similar experiments [3].
In future experiments, we will incorporate RNA-based systems within GUVs to model the implications of reoccurring content exchange in such prebiotic systems. In particular, we aim to combine freeze-thaw driven ribozyme activity with content mixing to probe if both processes could have synergistically driven primitive forms of protocell proliferation.

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P24 Could α-amino acid nucleotide copolymers play a role in early life?

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For decades views have been developed that life could have emerged from a single kind of biopolymers, namely RNA or peptides. These approaches are limited either by the availability of processes for the formation of RNA strands long enough to act as polymerase ribozymes or by the questionability of the replication of peptide sequences. Rather than having recourse to other kinds of monomers, we considered the possibility of mixed structures involving amino acid as well as nucleotide monomers bound by phosphoramidate and ester linkages. The results of experimental reactivity studies will be presented supporting the kinetic relevance of such structures as well as the availability of plausible chemical processes allowing their formation. Overall, they should not be considered as less probable than RNAs as contributors to the origin of life. The evolutionary advantage of these structures lies in the fact that the emergence of translation and the evolution of the ribosomal machinery can be understood as a logical outcome of their gradual improvement avoiding the need of a questionable transition from a pristine RNA world to an RNA-protein world in which the functional role is mostly carried out by proteins. An additional outcome of the reactivity investigation carried out during this work strongly suggest that translation should have evolved at low temperatures, close to 0°C in water or below in water-ice eutectic phases.

P25 Heated microbubbles condense and encapsulate molecules for early evolution

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Non-equilibrium mechanisms are crucial for the emergence of early life. They allow the fundamental continuous accumulation of prebiotic molecules into microcompartments. In addition, cyclic temperature changes, localized wet-dry cycles, crystallization, and encapsulation in lipids that undergo cell fission are considered crucial for the emergence of informational polymers. Here we show that all above mechanisms are triggered in less than 30 minutes at microbubbles in a heat flow. At the heated bubble, molecules accumulate by the continuous evaporation of water up to 1000-fold. The evaporating water recondenses at the cold side, implementing a heat pipe geometry that drives a continuous coffee-ring effect. The findings offer multiple reaction scenarios for the early chemistry of life. The setting is expected ubiquitously on early rocky planets where outgassing promotes gas bubbles, water is encapsulated in porous volcanic rock, and temperature gradients are provided from hydrothermal flows of steam or water.

P26 On the origins of the protein world: a computational approach to study the emergence of the first autonomously folding proteins

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Since the time of the Last Universal Common Ancestor (LUCA), proteins have been the fundamental catalysts of life. However, for their activity they must assume three-dimensional structures by a complex, easily disrupted, process of folding. Thus, it is still unclear how the first folded proteins emerged and how life came to rely so extensively on their ability to fold. Our hypothesis is that the first folded proteins resulted from the increased complexity of peptides in the "RNA-peptide world" that preceded LUCA, possibly by three mechanisms [1-2]: repetition, accretion, and recombination. By now, repetition and accretion have been explored with success, but recombination has so far remained poorly studied.
Due to its centrality to all processes of life and its very slow rate of change, the ribosome is the main survivor of the primordial "RNA-peptide world" and its proteins offer a window onto the time when polypeptide chains learned to fold [1]. Following a computational approach, we evaluated the propensity of different ribosomal fragments, which are intrinsically disordered and adopt regular structures only within a scaffold, to establish geometrically and energetically compatible interfaces that would allow the formation of stable, globular, recombinant folds in the absence of RNA. As a result, we identified more than 200 ribosomal fragment pairs that can recreate not only frequent protein folds but also novel fold topologies, opening a door to a better understanding of the emergence of the first autonomously folding proteins.

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P27 Data-driven astrochemistry: one step further within the origin of life puzzle

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Astrochemistry, meteoritics and chemical analytics represent a manifold scientific field, including various disciplines. Here, clarifications on astrochemistry, comet chemistry, laboratory astrophysics and meteoritic research with respect to organic and metalorganic chemistry will be given [1]. The seemingly large number of observed astrochemical molecules necessarily requires explanations on molecular complexity and chemical evolution, which will be discussed [2,3]. Special emphasis should be placed on data-driven analytical methods including ultrahigh-resolving instruments and their interplay with quantum chemical computations [4]. These methods enable remarkable insights into the complex chemical spaces that exist in meteorites and maximize the level of information on the huge astrochemical molecular diversity [5]. In addition, they allow one to study even yet undescribed chemistry as the one involving organomagnesium compounds in meteorites [3]. Both, targeted and non-targeted analytical strategies will be explained and may touch upon epistemological problems [1]. In addition, implications of (metal)organic matter toward prebiotic chemistry leading to the emergence of life will be presented [1]. The precise description of astrochemical organic and metalorganic matter as seeds for life and their interactions within various astrophysical environments may appear essential to further study questions regarding the emergence of life on a most fundamental level that is within the molecular world and its self-organization properties.

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P28 Exploring the role of geomaterials and magmatic activity on the emergence of life

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The melts, magma and experimental volcanology research group at LMU maintain a world-leading research capability in high temperature (up to 1600 °C) synthesis, characterization, and manipulation of geomaterials, very well equipped with a plethora of facilities to explore and characterize geomaterials and a variety of interactions involving them. These facilities include:
(1) high temperature gas-mixing furnaces, enhanced with a wide array of experimental manipulative technologies (e.g. chaotic mixing, buoyancy, rheology, splat quenching)
(2) a experimental facility for the manipulation of reaction kinetics between gas mixtures and geo-materials (e.g. glassy volcanic ash, mineral powders) in form of a high temperature tumbler-reactor. This facility enables the controlled development of reaction histories on the surface of powder grains.
(3) rapid decompression facilities for the explosive fragmentation and ejection of geomaterials from high temperature and high pressure (up to 30 MPa) into various atmospheres in the presence of lightning discharges. The same facility is also used to explore hydrothermal reactions and explosions.

The facilities are flanked by geochemical, petrological, petrophysical and mineralogical analyzing techniques to thoroughly characterize the geomaterials (minerals, rocks, ashes) prior and after experiments.

Such capabilities in experimental volcanology and petrology can provide invaluable insights to the research field of the emergence of life and foster new interdisciplinary experimental approaches; for instance (i) for the microfluidic replication chemistry by using various well-characterized geomaterials to explore how DNA behaves in the flow, or (ii) the experimental exploration of the fate, survival, and evolution of metalorganics in mineral and rock samples under conditions of precisely controlled temperature and oxygen fugacity, or (iii) the synthesis of prebiotic compounds under primordial volcanic conditions is being explored.

P29 Fischer-Tropsch reactions catalyzed by iron meteorites for prebiotic compounds

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The origin of life in the early Earth history necessitates the presence of key prebiotic organic compound. The source of this organic matter has been discussed intensively throughout the last decades. One prominent example is the experiment of Miller and Urey [1], which used lightning as a source of energy to convert gases like hydrogen, methane, ammonia, and water. Another experiment was performed by Studier [2,3], claiming that organic matter was formed from Fischer-Tropsch reactions in the solar nebula and subsequently transported to the early Earth via meteorites. Inspired by the latter, we investigate the possibility that Fischer-Tropsch reactions could also occur at Meteorite nanoparticles (formed during their entry into the Earth's atmosphere) on the hot early Earth, featuring a dense and reducing atmosphere. We, therefore, prepared metal nanoparticles with the metal ratio of two meteorites (Campo de Cielo and Muonionalusta) on two different supports (silica and montmorillonite clay) and used them as catalysts in Fischer-Tropsch reactions with H2 and CO2 varying the temperature, reaction time, pressure, and the H2:CO2 ratio. As a proof of concept, we additionally tested the meteorites themselves and their oxidized-reduced nanoparticles on montmorillonite clay as catalysts. In all experiments, we observe a significant formation of organic molecules such as n alkanes, iso alkanes, methanol, ethanol, toluene, and acetaldehyde. We conclude that Fischer-Tropsch reactions on meteorite nanoparticles occur under the conditions of the hot early Earth being a possible source of organic matter.

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P30 Monitoring the accumulation of molecules inside hydrothermal chambers via UV-Spectroscopy

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The accumulation of molecules inside porous rock of hydrothermal vents is of great interest for origin of life research [1]. Of special relevance is the behaviour of prebiotic molecules e.g. formamide (a potential precursor of nucleobases), single nucleobases themselves, and 1-ethyl-3-(3-dimethylaminopropyl)carbodiimide (EDC - a catalyst for polymerization and ligation reaction of phosphorylated nucleotides) [2-6]. Due to their size, the in situ observation of such small molecules inside microfluidic compartments is difficult. Using real time and specially resolved UV-spectroscopy the setup at hand allows for the quantification of molecules in aqueous solutions at micro- to millimolar concentrations. Preliminary results show the accumulation and hydrolysis dynamics of EDC and well as nucleobases inside mimicked hydrothermal pores. The setup opens new perspectives in quantifying and observing processes such as EDC polymerization reactions of single nucleotides. Moreover, we can monitor chemical conversions, thermophoresis, and non-equilibria of chemical gradients (e.g. concentrations, redox and pH gradients) directly or via gradient sensing molecular systems, going beyond the fluorescence-based methods used previously [7].

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P31 Novel insights into enantiomeric enrichment and their role in prebiotic peptide synthesis

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Evolution of homochirality from an achiral world is key for understanding the concept of life. Even though some possibilities for the shift out of racemic state have been published, we are still in search for systems that also amplify chirality [1]. The work of Kagan and Agami about non-linear effects as well as Soai on self-amplifying systems allow a first impression on possible mechanisms leading to a homochiral world [2, 3]. Transferring these basic principles to a prebiotic world setup has become a focus of interest. In this regard, saturation effects and preferential crystallization of racemates and conglomerates play an important role in enantio¬enrichment and enantiodepletion of amino acids. The search for enrichment factors and implementing the most promising in prebiotic peptide formation in highly complex amino acid mixtures promises insight into the emergence of homochirality on early earth.

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P32 Efficient prebiotic self-repair of DNA photolesions involving purine nucleobase analogs

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Cyclobutane pyrimidine dimers (CpDs) are considered as the most common DNA lesions arising during the exposure of the biopolymer to UV light. While these lesions can be repaired in living organisms by e.g. photolyases, such sophisticated repairing factors were absent on Archean Earth. Recent experimental findings suggested that specific DNA sequences exhibit fascinating self-repairing capabilities, which could efficiently protect them from the detrimental effects of pyrimidine dimerization [1]. Nevertheless, the exact molecular mechanism of this process remained obscure until recently, owing to considerable challenges associated with synthesis of selectively damaged sequences. Here, I will describe the mechanistic details of the self-repair process determined by means of MD simulations and highly accurate quantum-chemical calculations involving the algebraic diagrammatic construction to the second order method, ADC(2) [2]. In particular, the most recent results revealed that the UV-induced self-repair of the GAT=T sequence is triggered by sequential electron transfer process operating downhill along the slope of the potential energy surface in the lowest excited singlet state. The understanding of the mechanistic details of this process enables us to predict not only the most efficient self-repairing DNA sequences, but also select the most prominent alternative nucleobases which conduct the cleavage of CpDs even more effectively than their canonical counterparts.

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P33 Investigation of nucleotides as prebiotic plausible organocatalysts and their application in formose-type reactions

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In the content of origin of life, the RNA World hypothesis has been proposed [1]. RNA is already attributed with evolutionary properties such as information storage, catalytic activity, further development through mutation and independent replication [2]. The formation of RNA and its components such as sugars, nucleobases and nucleotides has already been demonstrated under prebiotic conditions [3]. Under the assumption of independent replication, we want to investigate the catalytic influence of nucleotide monophosphates (NMP´s) on the formose reaction. In particular, we show the influence of different cations on the catalytic activity of NMP´s. Furthermore, we investigate the effect of different NMPs on the selectivity of the formation of certain sugars during the formose reaction.

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P34 Dissipative assemblies that inhibit their deactivation

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Inspired by biological dissipative assemblies, such as the GTP driven formation of microtubules, researchers have started exploring this non-equilibrium approach to develop complex systems aiming to mimic the properties found on their biological counterparts.[1]
In our lab we couple the assembly process to chemical reaction cycles which consume fuel driving as a result, the assemblies out of equilibrium. More precisely, carboxylic acid precursors are transformed into their corresponding anhydride products by consumption of chemical fuels as carbodiimides. The product is metastable and rapidly hydrolyzes back to the precursor. When the product concentration exceeds its solubility value, different assemblies are formed including colloids or droplets.[2]
We study in detail the underlying mechanism of the inhibition of the hydrolysis reaction in the presence of these assemblies. We show that the mechanism is a result of the assemblies protecting its anhydride product from deactivation. We demonstrate that we can tune the level of the inhibition of the hydrolysis. With that control, we can manipulate the survival time of the assemblies and their persistence towards periods of starvation from their energy source. [3]

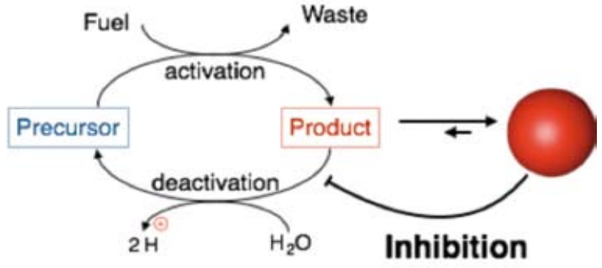

Figure 1 – Scheme of the inhibition mechanism containing the chemical reaction coupled to assemblies

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P35 Towards synthetic cells by using peptide-based reaction compartments

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Growth and division of a life like compartment require production or external supply of membrane components, which is coupled to their internal chemical dynamics. These components are amphiphilic molecules such as fatty acids, phospholipids or amphiphilic peptides that create membranous enclosures via self-assembly [1-3]. In order to synthesize lipids inside of such protocells de novo, complex enzymatic pathways have to be installed [4]. By contrast, peptides can be generated more easily by in vitro gene expression from a genetic template. Here, we use amphiphilic elastin-like polypeptides (ELP) [5] to create self-assembled vesicular structures of ≈200 nm diameter as a possible path towards a synthetic cell. We then demonstrate encapsulation of cell-free transcription and translation reactions inside the peptide vesicles via compartmentalized production of fluorescent RNA aptamers and proteins. We finally, succeed in the expression of the membrane-constituting peptides inside the vesicles themselves and demonstrate their incorporation into the membrane and thus inherent vesicle growth.

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P36 Formation of temporary vesicles using dissipative self-assembly

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Dissipative molecular self-assembly enables researchers to develop complex systems that exist out of equilibrium. Those systems resemble their biological counterparts, like the GTP-driven formation of microtubules. [1] Another important molecular assembly, especially in biological cells, are vesicles, since they are semi-permeable and ideal to compartmentalize.
In our laboratory we established a chemical reaction network based on water-soluble dicarboxylate precursors that intramolecular react to their corresponding anhydrides using carbodiimides as a chemical fuel. [2] The increased hydrophobicity induces the out of equilibrium self-assembly into various morphologies like fibers [2], oil-droplets [3], colloids [2,4] and vesicles. The anhydride product rapidly hydrolyzes back to its precursor state leading to the degradation of the assemblies. By changing the precursor, we are able to vary the nature of the assembly, while with the amount of added fuel the lifetime can be changed drastically from minutes to hours.
Here we report the formation of dissipative vesicles using the aforementioned reaction network and extremely simple tripeptide or aliphatic precursors. Through the different backbones and their unique interactions, we want to investigate the kinetic mechanism to approach size control or even more complex behavior like self-replication of the vesicles.

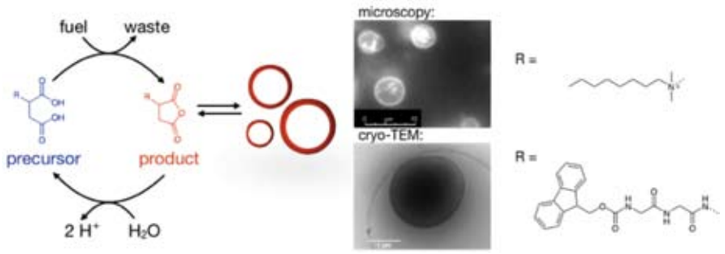

Fig. 1:
Reaction network and precursors to form dissipative vesicles with images from light microscopy and cryo-TEM.

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P37 The prevalence of conservative evolution in the protein sequence universe

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The genesis of a structured and functional protein by random processes is exceedingly unlikely. However, once a functioning protein emerges, it can easily gain acceptance [1]. The evolution of natural proteins therefore often proceeds through the amplification of already established protein sequences. Copies of the same sequence evolve over time, leading to the co-existence of similar sequences that might also have diversified in function [2].
We investigate the prevalence of such conservative evolution by analyzing reuse in the protein sequence universe. 1300 non-redundant bacterial genomes of distinct genera with exemplars from most bacterial classes are chosen as a representative for this study. We use statistical modeling in order to distinguish sequence similarities arising through reuse, as opposed to mere chance. For this purpose we derive the distribution of point mutation distances between randomly drawn k-mers. For long point mutation distances, the distribution can be described by a binomial distribution based on the amino acid composition of the underlying data. The frequency of shorter distances is significantly increased relative to the binomial distribution and can be explained by reuse.
In the example of 100mers, we find that most sequence fragments (>90%) are at least reused once (p-value of 10-5). More than 10% of all sequence fragments are extensively reused and reoccur more than thousand times. Pairwise genome comparison reveals an overlap of around 19% common sequences on average. This demonstrates that the pressure to conserve sequences is strong enough to cause such significant sequence overlap, even after billions of years have passed.

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P38 Role of RNA/DNA bases in primordial photosynthesis

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Hard ultraviolet (UV) radiation was abundant on earth at the beginning of life. Since the RNA and DNA bases are efficient UV absorbers, the photophysical properties of the bases, nucleosides and nucleotides must have played a crucial role for the origin of life. It is nowadays widely accepted that exceptional UV-photostability is an essential property which led to the selection of RNA/DNA bases at the origin of life [1,2]. While the energy of UV-excited electronic states is potentially harmful, leading to the destruction of organic chromophores, this energy is rapidly converted into heat in RNA/DNA bases by radiationless transitions through specific conical intersections of the electronic potential-energy surfaces [3] and the heat is subsequently dissipated into the environment. This discussion [1-3] has overlooked that photoexcited RNA/DNA bases react readily with water by abstracting hydrogen atoms from water or ejecting hydrogen atoms into water, thus decomposing water into free H and OH radicals. By state-of-the-art quantum-chemical computations we have explored these reaction mechanisms for the example of adenine in a microscopic water environment [4,5]. In the first photochemical step, adenine (A) abstracts an H-atom from a hydrogen-bonded water molecule which yields AH and OH radicals. In the second photochemical step, the H-atom is photodetached from the AH radical, which yields a free H-atom and regenerates A, which thus becomes a photocatalyst. Overall, a water molecule hydrogen-bonded to A is thus decomposed into free H and OH radicals. Adenine in an aqueous environment thus is UV-photostable, but water is not. Adenine is a photocatalyst for the homolytic decomposition of water. The free H radicals are strongly reducing agents which can initiate the reduction of carbon dioxide, which may have been the beginning of photosynthesis.

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