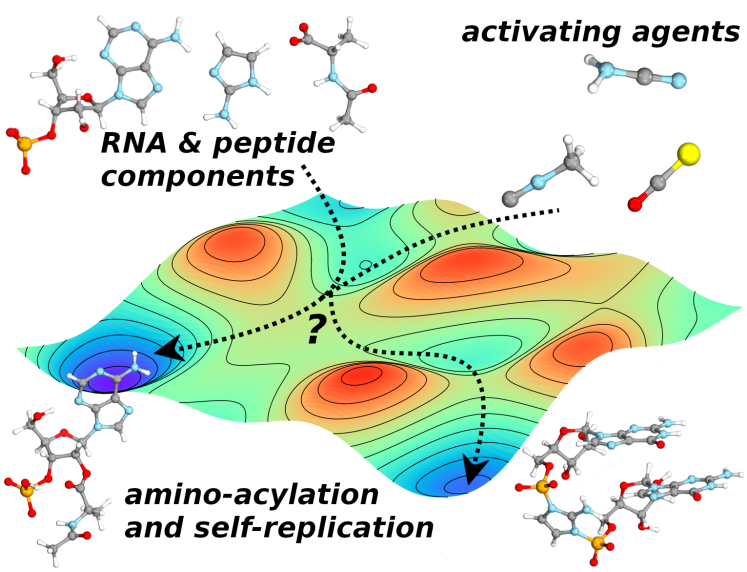
This project will provide molecular understanding about the way prebiotic mixtures of bioorganic molecules could harness chemical energy to evolve towards complexity and life-like chemical processes. Living organisms require a constant supply of energy to maintain their primary functions, such as copying of the genetic material or protein synthesis. Cells achieve this for example through the decomposition of sugars and stores the acquired energy in the form of adenosine triphosphate (ATP), the universal energy currency of life. However, this is only possible with the support of complex enzymatic machinery, which was absent at the advent of life. Therefore, recent scenarios for the early minimal metabolism considered more reactive, albeit less stable, high-energy organic molecules with high energy triple bonds. Nevertheless, these energy-storing and activating molecules were shown to suffer from low selectivity and high propensity to udergo hydrolysis-based degradation. Most importantly, their mechanism of action as well as the underlying thermodynamics and kinetics remain largely obscure and detailed studies at the molecular level are necessary to understand what form of minimal metabolism could fuel the early molecular evolution.



In this project, I will employ accurate methods of quantum chemistry to investigate the molecular mechanisms of action of such high-energy molecules as well as prebiotically relevant reactions that could enable their formation. Here, computational chemistry will serve as a molecular microscope which will allow to uncover the inner workings of prebiotic energy harnessing and storage. In particular, (1) we will investigate what sort of prebiotic molecular fuel could sustain self-replication of informational polymers such as RNA and the synthesis of peptides. By constructing chemical reaction networks and simulating their operation over time, we will learn what modifications of the network could affect the product ratios and selectivity. (2) We will also elucidate the formation of aminonucleoside amido-triphosphates under prebiotic conditions and investigate their propensity to undergo polymerisation. Such triphosphosphorylated analogs of RNA monomers were recently shown as prebiotically attainable forms active building blocks which could potentially enable self-replication of the genetic material. Finally, (3) we will elucidate the chemical routes to high-energy molecules, containing cyanide groups, in the interstellar media. In this case, we will investigate photoinduced processes and radical reactions that are responsible for the formation of such complex organic molecules under astrochemical conditions.

The results of these computational efforts will allow us to establish general principles governing the utilization and storage of chemical energy by prebiotic systems. Elucidating such principles based on thermodynamics, kinetics and molecular mechanisms will make it easier to understand the chemistry that could give rise to primitive forms of life in the universe and offer stronger hypotheses about potential biosignatures in the Solar system and beyond. Such insights will be particularly valuable and timely considering the recent launches of the Perseverance rover and the James Webb Space Telescope as well as the planned DAVINCI mission to Venus, all of which aim to explore the types of chemistry could give rise to life beyond Earth.