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Can a reaction's environment program its outcome, and does it matter?

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Chemists go to great lengths to avoid making complex mixtures:[1] because they expect molecules of interest to be so dilute as to result in non-functional systems ('tar') [2,3] and because they are expected to be 'analytically intractable'.[4,5] This is often especially true in Origins research,[6] where the taming of combinatorial explosion in 'uncontrolled' condensation reactions of bio-monomers or their analogues is an important open question. Controlling chemical complexity generated in condensation reactions is enormously challenging, and currently only achieved using strategies such as protecting group chemistry to produce single defined products.[1]

Our group has been exploring the complexity generated by 'uncontrolled' condensation reactions of polyvalent monomers, which most chemists eschew. We are interested in how it can be tamed by the recursive interaction with environments, and how complex ensembles of products may have consistently defined structural and functional properties (even if their composition is not comprehensively understood).[7] This is a 'systems' approach to reaction complexity (set out Figure 1), in contrast to planned organic syntheses/disconnections. As model systems, we have studied the condensation of both simple pure building blocks[8] and complex mixtures of the kind produced by prebiotic monomer syntheses.[4, 6] We have used analytical approaches more common to Systems Biology to take a new approach to the synthesis of functional macromolecules, and are combining these with inspection of structure and function. In this contribution I will present our latest results,[6] demonstrating the impact of an environments ability to steer product distribution in complex reaction systems, leading to real and reproducible effects in the realms of structure and function.

References: [1] Merrifield, RB (1985) *Angewandte Chemie, International Edition* 24: 799-810. [2] Benner, SA, Kim, HJ & Carrigan, MA (2012) *Accounts of Chemical Research* 45: 2025-2034. [3] Schuster, P (2000) *Proceedings of the National Academy of Sciences* 97:7678-7680. [4] Schwartz, AW (2007) *Chemistry & Biodiversity* 4:656-664. [5] Bissette, A (2013) *Nature Chemistry* 5: 729. [6] Ruiz-Mirazo, K (2014) *Chemical Reviews* 114:285-366. [7] Surman, AJ et. al. (2017) *In Preparation* [8] Rodriguez-Garcia M (2015) *Nature Communications* 6:8385.

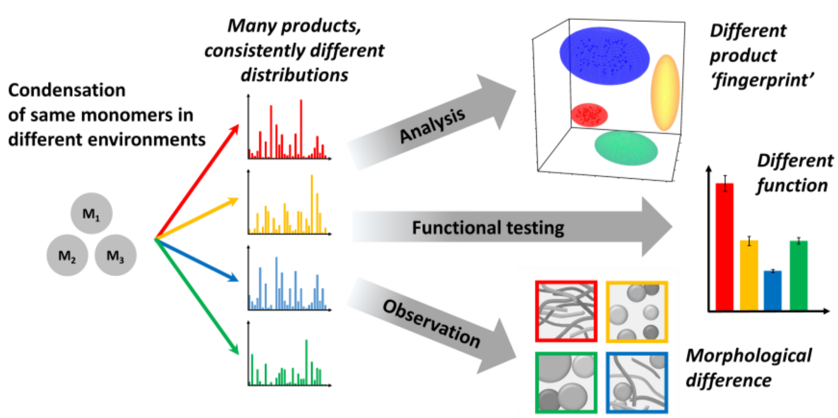

Figure 1 – Center figures and captions after the text of the abstract.