

A Lanthipeptide-like N-Terminal Leader Region Guides Peptide Epimerization by Radical SAM Epimerases: Implications for RiPP Evolution

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Abstract

Ribosomally synthesized and posttranslationally modified peptide natural products (RiPPs) exhibit diverse structures and bioactivities and are classified into distinct biosynthetic families. A recently reported family is the proteusins, with the prototype members polytheonamides being generated by almost 50 maturation steps, including introduction of d-residues at multiple positions by an unusual radical SAM epimerase. A region in the protein-like N-terminal leader of proteusin precursors is identified that is crucial for epimerization. It resembles a precursor motif previously shown to mediate interaction in thioether bridge-formation in class I lanthipeptide biosynthesis. Beyond this region, similarities were identified between proteusin and further RiPP families, including class I lanthipeptides. The data suggest that common leader features guide distinct maturation types and that nitrile hydratase-like enzymes are ancestors of several RiPP classes.

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