Total synthesis and structural validation of cyclodepsipeptides solonamide A and B

Microorganisms are an attractive source of new natural products with antimicrobial properties, and the marine environment constitutes a prolific resource of bioactive microorganisms. During a global research expedition (Galathea III), two depsipeptides, solonamide A and solonamide B, were isolated from the marine bacterium Photobacterium halotolerance and were found to inhibit virulence gene expression in the serious human pathogen, Staphylococcus aureus. They act by interfering with the agr quorum sensing system and show resemblance to the endogenous S. aureus quorum sensing peptide, autoinducing peptide I (AIP-I). To enable more comprehensive studies, we embarked on the chemical synthesis of solonamides A and B. The key synthetic steps were formation of the (R)-β-hydroxy-fatty-acids by stereo-selective aldol reactions and a cyclative macrolactamization, which proceeded under highly dilute conditions. Thus, the first total syntheses of the solonamides corroborated the originally assigned structures, and by changing the stereochemistry of the auxiliary in the aldol steps we gained access to the natural products as well as their β3-epimers.

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