

The boat-shaped polyketide resistoflavin results from re-facial central hydroxylation of the discoid metabolite resistomycin.

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Abstract

Resistoflavin (1) is a rare boat-shaped pentacyclic polyketide metabolite of *Streptomyces resistomycificus* with marked antibacterial activity. By a series of experiments we have disclosed that the optically active molecule is derived from the discoid polyketide resistomycin (2) by an unusual, enantioface-differentiating hydroxylation, which leads to the capped pentacyclic ring system. In vivo and in vitro experiments unequivocally demonstrate that this reaction is catalyzed by RemO, an FAD-dependent monooxygenase. In addition, we were able to establish the absolute configuration of 1 and thus the stereochemical course of this rare enzymatic reaction by extensive computational methods. Comparison of the experimental CD spectrum with those quantum chemically calculated for (R)-1 and (S)-1 revealed the R-configuration of 1. Consequently, the enzyme-catalyzed hydroxylation takes place from the Re-face of 2 with loss of aromaticity in favor of a chiral carbinol center. While other oxygenases involved in polyketide tailoring functionalize the periphery of polyphenols, RemO is unique in its ability to catalyze a central, nonperipheral

hydroxylation of a fused ring system.

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