

Assembly of melleolide antibiotics involves a polyketide synthase with cross-coupling activity.

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Abstract

Little is known about polyketide biosynthesis in mushrooms (basidiomycota). In this study, we investigated the iterative type I polyketide synthase (PKS) ArmB of the tree pathogen *Armillaria mellea*, a producer of cytotoxic melleolides (i.e., polyketides esterified with various sesquiterpene alcohols). Heterologously produced ArmB showed orsellinic acid (OA) synthase activity in vitro. Further, we demonstrate cross-coupling activity of ArmB, which forms OA esters with various alcohols. Using a tricyclic *Armillaria* sesquiterpene alcohol, we reconstituted the biosynthesis of melledonol. Intermolecular transesterification reactions may represent a general mechanism of fungal PKSs to create structural diversity of small molecules. Phylogenetic network construction of thioesterase domains of both basidiomycetes and ascomycetes suggests that the fungal nonreducing PKS family has likely evolved from an ancient OA synthase and has gained versatility by adopting Claisen-like cyclase or transferase activity.

Involved units

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