Unexpected Metabolic Versatility in a Combined Fungal Fomannoxin/Vibralactone Biosynthesis.

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Abstract

The secondary metabolome of an undescribed stereaceous basidiomycete (BY1) was investigated for bioactive compounds. Along with a known fomannoxin derivative and two known vibralactones, we here describe three new compounds of these natural product families, whose structures were elucidated using 1D and 2D NMR spectroscopy and high-resolution mass spectrometry. The new compound vibralactone S (4) shows a 3,6-substituted oxepin-2(7H)-one ring system, which is unprecedented for the vibralactone/fomannoxin class of compounds. Stable isotope labeling established a biosynthetic route that is dissimilar to the two published cascades of oxepinone formation. Another new compound, the antifungal methyl seco-fomannoxinate (6), features a 2-methylprop-1-enyl ether moiety, which is only rarely observed with natural products. The structure of 6 was confirmed by total synthesis. (13)C-labeling experiments revealed that the unusual 2-methylprop-1-enyl ether residue derives from an isoprene unit. The diversity of BY1's combined fomannoxin/vibralactone metabolism is remarkable in that these compound families, although biosynthetically related, usually occur in different organisms.

Identifier

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