



Studies in Natural Products Chemistry: Chapter 5. Abyssomicins: Isolation, Properties, and Synthesis

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The discovery of abyssomicin class of natural products has been the outcome of the search for secondary metabolites possessing inhibitory effect on p-aminobenzoic (pABA) acid biosynthesis, a biosynthetic pathway found in many microorganisms but not in humans. The earliest members of this family, abyssomicins B, C, and D, were isolated from the marine actinomycete *Verrucosispora AB-18-032*, collected from a sediment sample from Japanese sea at 289m depth. Abyssomicin C was the first natural product to act as an inhibitor of the enzymes involved in pABA and folic acid biosynthesis. In addition to these initial members of the family, other related compounds were also isolated and characterized: atrop-abyssomicin C, abyssomicins G, H, E, I, J, K, and L. Interestingly, only abyssomicin C and atrop-abyssomicin C, which was initially discovered during the synthesis of the former compound and later isolated from the fermentation product, show activity against Gram-positive bacteria. The structural resemblance of abyssomicins to the chorismic acid transition state analogues is evident, while distinctive activity is related to the presence of the conjugated ketone functionality. The structural novelty of abyssomicins and very promising biological properties spurred interest among synthetic organic chemists. The complex molecular architecture and a number of challenging structural elements, such as a fused tetronate bicyclic core with a strained 11-membered macrocyclic ring, necessitate a conscientious synthetic plan and several routes were reported in the literature. Intramolecular Diels–Alder reaction was used in the biomimetic synthesis as a strategically major step in building the polycyclic skeleton (below scheme, path A). The diene functionality was attached to the tetronic moiety possessing the exocyclic double bond to set a scene for the key cycloaddition step, which then afforded the core abyssomicin structure in a highly regio- and diastereoselective manner. Further elaboration of the cycloadduct produced the abyssomicin C. Related but different strategy employed the Lewis acid-templated intermolecular Diels–Alder reaction to assemble a highly substituted cyclohexane product (above scheme, path B). The cyclohexane ring formation via the cycloaddition reaction was followed by introduction of the tetronic moiety to form the spiro compound. The macrocyclic ring was then introduced by combining the nucleophilic properties of 2-Li-tetronate and the ring-closing metathesis as key transformations. An alternative approach to the Diels–Alder reaction employed an intramolecular variant of the nucleophilic displacement onto the π -allyl palladium complex to afford a highly substituted cyclohexane derivative (above scheme, path C). Further elaboration of this product provided access to the spirotetronic derivative via an attractive gold-catalyzed cyclization of the propargyl compound. Final assembly of the macrocyclic ring was accomplished employing the nucleophilic properties of 2-Li-tetronate and, in this instance, a particularly impressive Nozaki–Hiyama–Kishi reaction.



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