Strategies and Tactics in Organic Synthesis: Chapter 4. Total Synthesis of (±)-Anislactone A and (±)-Merrilactone A

Michael F. Greaney, Lei Shi, Naim Nazef

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This chapter describes the synthesis of two related sesquiterpenes, anislactone A and merrilactone A. We initially accessed a tetracyclic oxetane in the merrilactone series using a Paternò–Büchi reaction but found the compound to be too underfunctionalized to advance further. We then developed an approach based on reductive epoxide ring opening, whereby a fully elaborated C-ring epoxy-cyclopentane, containing five stereocenters, could undergo reductive epoxide cleavage when treated with Ti(III). The resulting tertiary radical then participates in a 5-exo-dig cyclization onto a pendant alkyne to afford the complete carbon skeleton of both natural products. From this point, orthogonal functionalization routes enabled the synthesis of both anislactone A and merrilactone A. A second-generation merrilactone A synthesis is then described, growing out of discoveries made over the course of the first route in the area of cyclopentannulation. An iodo-aldol method was used to develop an approach to the anislactone skeleton and succeeded in producing the BC bicycle with good stereocontrol and functional group tolerance. Further functionalization, however, did not prove possible due to excessive steric hindrance around the incorporated iodo group preventing any productive transformation. This problem was solved by switching the nucleophile in the tandem-aldol process to cyanide. The resulting domino cyanide-addition aldol cyclization was then successfully employed in the formal synthesis of merrilactone A, using a late-stage [2+2] photocycloaddition to access the D-ring.

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