Supporting Information
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Supporting Information
Asymmetric organocatalytic Michael addition-cyclization cascade of cyclopentane-1,2-dione with substituted α,β-unsaturated aldehydes

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X-ray structure

Crystal data for 3c:

C_{14}H_{13}ClO_3, M_r = 264.69, Monoclinic, P2_1, a = 8.1515(2) Å, b = 8.9479(2) Å, c = 35.9124(7) Å, β = 91.5626(12)°, V = 2618.43(10) Å³, Z = 8, Mo-Kα radiation (λ = 0.71073 Å) at T = 170.0(1) K, 46933 reflections measured, of which 10922 unique (9055 with I > 2σ(I)), R_{int} = 0.042, R_1[F^2 > 2σ(F^2)] = 0.041, wR^2 (all data) = 0.091, S = 1.01, Flack x = 0.076(18); absolute structure determined by anomalous diffraction effects using 3559 quotients [(F^+)-(F^-)]/[(F^+)+(F^-)]. The crystallographic data is deposited with the Cambridge Crystallographic Data Centre (CCDC 1528060) and can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.

Experimental section

Single crystals of the compound 3c were obtained by slow diffusion of hexane into a solution of 3c in EtOAc. Small colorless needle-like crystals were formed after two days. Single crystal X-ray diffraction data was collected at 170K on a Bruker-Nonius KappaCCD diffractometer, equipped with a detector, using graphite-monochromatized Mo-Kα radiation (0.71073Å). COLLECT was used for data collection. DENZO and SCALEPACK were used for data reduction and SADABS 2008 for absorption correction. The structure was solved using SHELXT and refined by full-matrix least-squares method against F^2 with SHELXL-2014 through the OLEX2 program package. All non-hydrogen atoms were refined with anisotropic atomic displacement parameters, hydrogen atoms were treated as riding on their parent carbon and oxygen atoms, with U_{iso}(H) = 1.2U_{iso}(C) for CH and CH_2, U_{iso}(H) = 1.5U_{iso}(C) for CH_3 and OH. The compound was found to crystallize in a non-centrosymmetric space group P2_1, with eight molecules of 3c in the unit cell. The absolute configuration of the compound was determined from the anomalous dispersion of the chlorine atoms to be (S,S). The figures were drawn using the program Mercury CSD 3.3.

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$^1$H and $^{13}$C NMR spectra
\((2S,4S)-2\text{-hydroxy-4-phenyl-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one} \) 3a
(2S,4S)-2-hydroxy-4-(4-methoxyphenyl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3b
(2S,4S)-4-(4-chlorophenyl)-2-hydroxy-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3c
(2S,4S)-4-(4-bromophenyl)-2-hydroxy-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3d
(2S,4S)-2-hydroxy-4-(4-nitrophenyl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3e
(2S,4R)-2-hydroxy-4-(2-nitrophenyl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3f
(2S,4R)-4-(furan-2-yl)-2-hydroxy-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3g
(2S,4R)-4-butyl-2-hydroxy-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3h
(2S,4S)-2-hydroxy-4-((E)-prop-1-en-1-yl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3i
(S)-2-hydroxy-3-(3-hydroxy-1-phenylpropyl)cyclopent-2-en-1-one 5a
(S)-2-hydroxy-3-(3-hydroxy-1-(4-methoxyphenyl)propyl)cyclopent-2-en-1-one 5b
(S)-3-(1-(4-chlorophenyl)-3-hydroxypropyl)-2-hydroxycyclopent-2-en-1-one 5c
(S)-3-(1-(4-bromophenyl)-3-hydroxypropyl)-2-hydroxycyclopent-2-en-1-one 5d
(S)-2-hydroxy-3-(3-hydroxy-1-(4-nitrophenyl)propyl)cyclopent-2-en-1-one 5e
(S)-2-hydroxy-3-(3-hydroxy-1-(2-nitrophenyl)propyl)cyclopent-2-en-1-one 5f
(R)-3-(1-(furan-2-yl)-3-hydroxypropyl)-2-hydroxycyclopent-2-en-1-one 5g
(R)-2-hydroxy-3-(1-hydroxyheptan-3-y1)cyclopent-2-en-1-one 5h
(S,E)-2-hydroxy-3-(1-hydroxyhex-4-en-3-yl)cyclopent-2-en-1-one 5i
4-phenyl-3,4,5,6-tetrahydrcyclopenta[b]pyran-7(2H)-one 6
HPLC Chromatograms
(2S,4S)-2-hydroxy-4-phenyl-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one rac-3a
(2S,4S)-2-hydroxy-4-phenyl-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3a
(2S,4S)-2-hydroxy-4-(4-methoxyphenyl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one rac-3b
(2S,4S)-2-hydroxy-4-(4-methoxyphenyl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3b

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(2S,4S)-4-(4-chlorophenyl)-2-hydroxy-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3c
(2S,4S)-4-(4-bromophenyl)-2-hydroxy-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one rac-3d
(2S,4S)-4-(4-bromophenyl)-2-hydroxy-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3d
(2S,4S)-2-hydroxy-4-(4-nitrophenyl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one rac-3e
(2S,4S)-2-hydroxy-4-(4-nitrophenyl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3e
(2S,4S)-2-hydroxy-4-(2-nitrophenyl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one rac-3f
(2S,4S)-2-hydroxy-4-(2-nitrophenyl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3f
(2S,4R)-4-(furan-2-yl)-2-hydroxy-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one rac-3g
(2S,4R)-4-(furan-2-yl)-2-hydroxy-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3g
(2S,4R)-4-butyl-2-hydroxy-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one rac-3h
(2S,4R)-4-butyl-2-hydroxy-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3h
(2S,4S)-2-hydroxy-4-((E)-prop-1-en-1-yl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one rac-3i
(2S,4S)-2-hydroxy-4-((E)-prop-1-en-1-yl)-3,4,5,6-tetrahydrocyclopenta[b]pyran-7(2H)-one 3i
(S)-2-hydroxy-3-(3-hydroxy-1-phenylpropyl)cyclopent-2-en-1-one 5a
(S)-2-hydroxy-3-(3-hydroxy-1-(4-methoxyphenyl)propyl)cyclopent-2-en-1-one 5b
(S)-3-(1-(4-chlorophenyl)-3-hydroxypropyl)-2-hydroxycyclopent-2-en-1-one 5c
(S)-3-(1-(4-bromophenyl)-3-hydroxypropyl)-2-hydroxcyclopent-2-en-1-one 5d
(S)-2-hydroxy-3-(3-hydroxy-1-(4-nitrophenyl)propyl)cyclopent-2-en-1-one 5e
(S)-2-hydroxy-3-(3-hydroxy-1-(2-nitrophenyl)propyl)cyclopent-2-en-1-one 5f

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Totals: 5.82577e4 1750.25617
(R)-3-(1-(furan-2-yl)-3-hydroxypropyl)-2-hydroxycyclopent-2-ene-1-one 5g
(R)-2-hydroxy-3-(1-hydroxyheptan-3-yl)cyclopent-2-en-1-one 5h

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(S,E)-2-hydroxy-3-(1-hydroxyhex-4-en-3-yl)cyclopent-2-en-1-one 5i