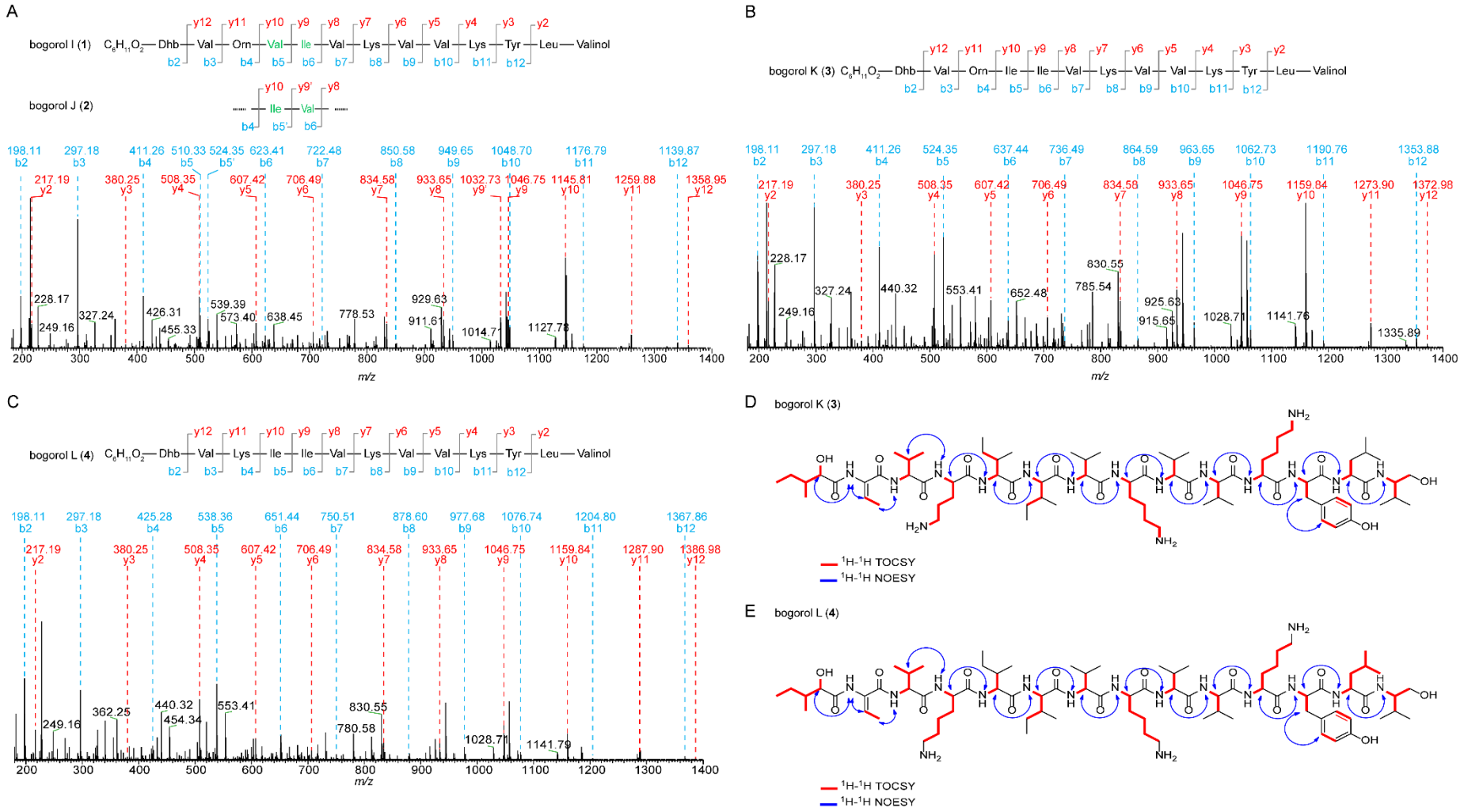


Fig. S1. LC-MS/MS and NMR analysis of bogorol I-L(**1**-**4**). Fragmentation of bogorol I and J (A), bogorol K (B), and bogorol L (C) by tandem MS to generate *b* and *y* ions. The *b* ions were indicated in blue, while the *y* ions were indicated in red. The different amino acids of bogorol I and bogorol J are indicated in green. ^{1}H-^{1}H TOCSY and ^{1}H-^{1}H NOESY NMR correlations of bogorol K (D) and bogorol L (E).

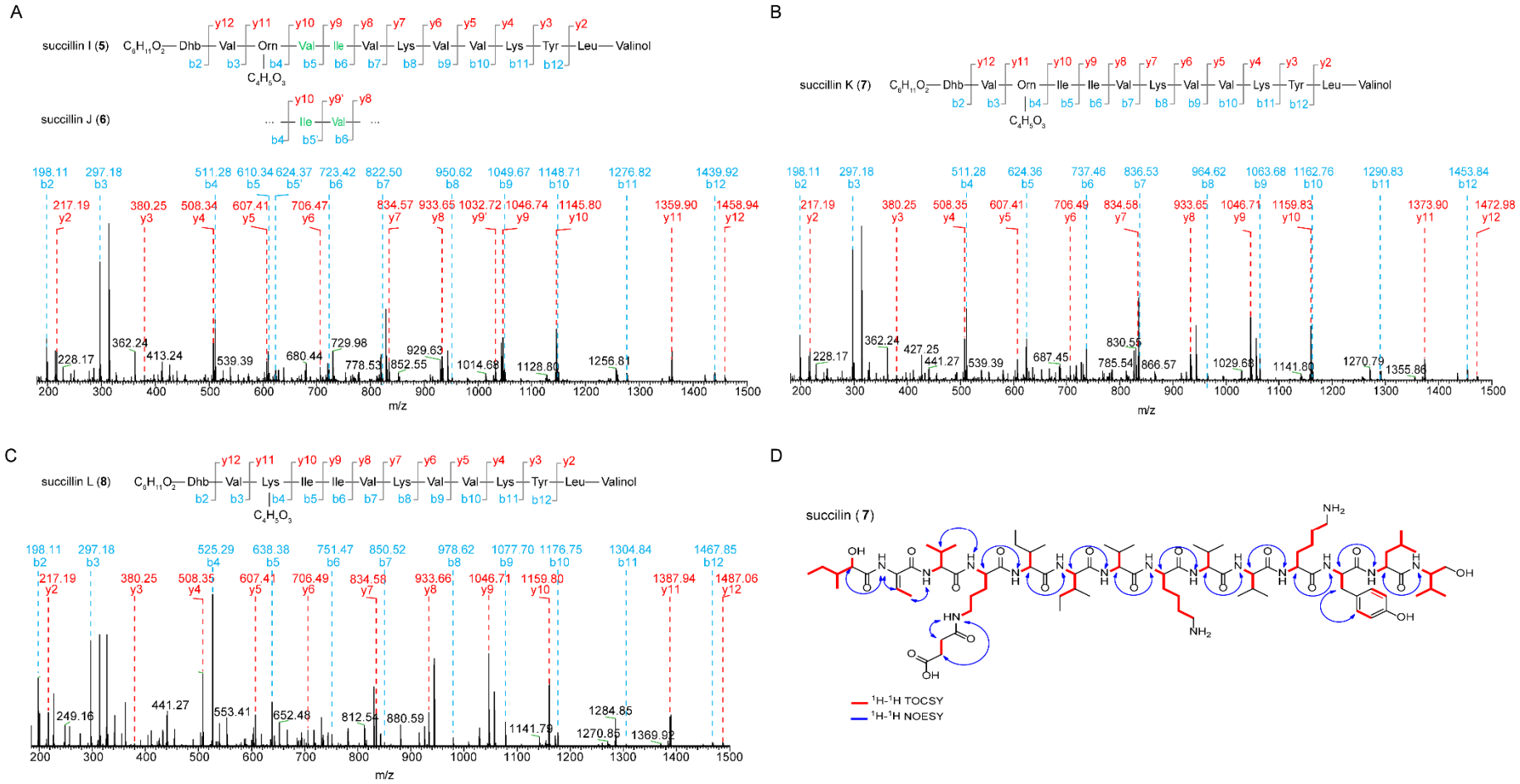


Fig. S2. LC-MS/MS and NMR analysis of succilin I-L(**5**-**8**). Fragmentation of succilin I and J (A), succilin K (B), and succilin L (C) by tandem MS to generate *b* and *y* ions. The *b* ions were indicated in blue, while the *y* ions were indicated in red. The different amino acids of succilin I and succilin J are indicated in green. ^{1}H-^{1}H TOCSY and ^{1}H-^{1}H NOESY NMR correlations of succilin K (D)

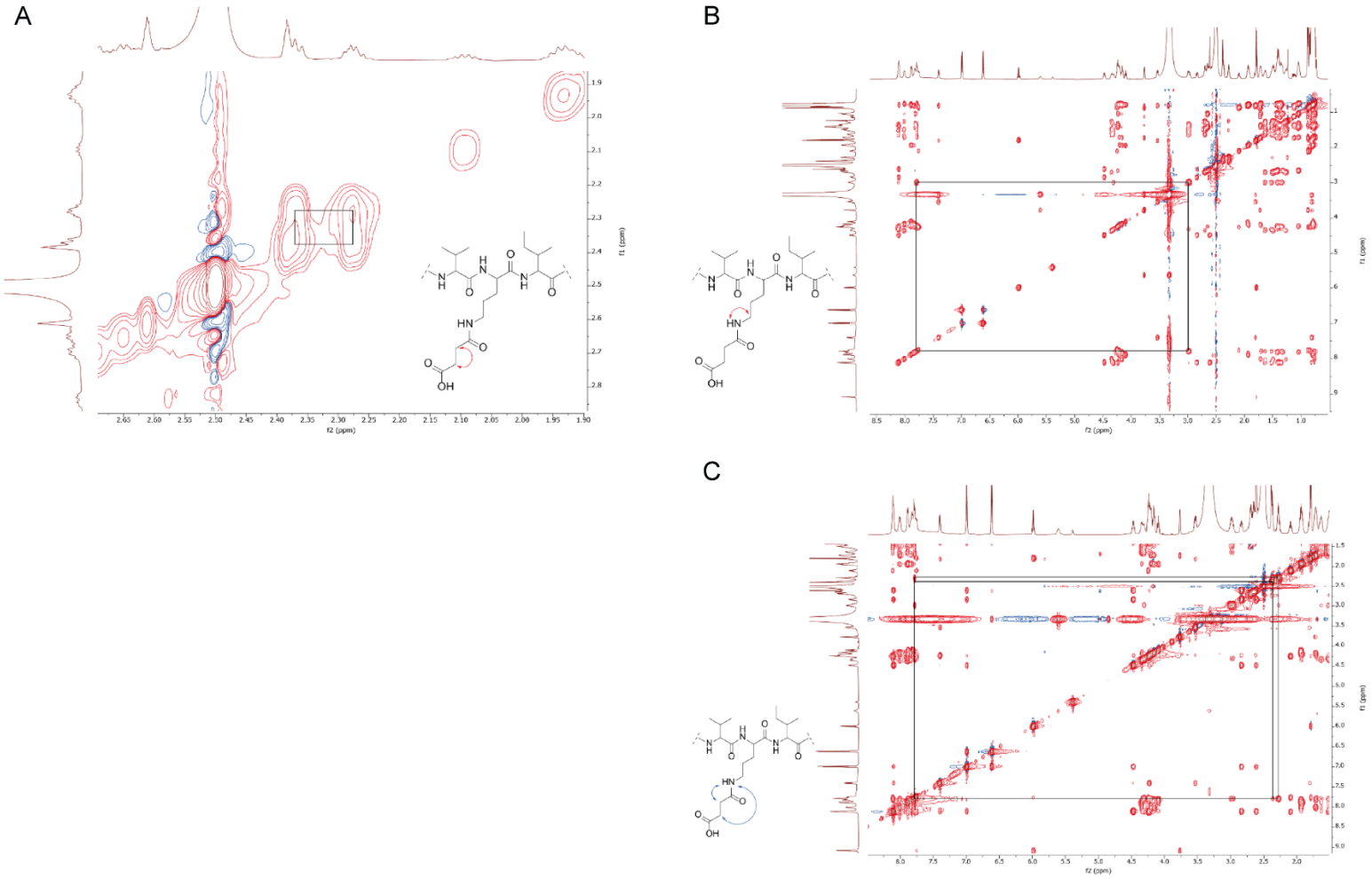


Fig. S3.NMR analysis of succilin K (**7**) to verified the succinylation. (A) ^{1}H-^{1}H-TOCSY NMR correlation of the delta methylene of Orn_3 with the amide NH of the succinyl group of succilin K. (B) ^{1}H-^{1}H-TOCSY NMR correlation of the delta methylene of Orn_3 with the amide NH of the succinyl group of succilin K. (C) ^{1}H-^{1}H-NOESY NMR correlations of the succinyl methylenes with the amide NH of the succinyl group of succilin K.

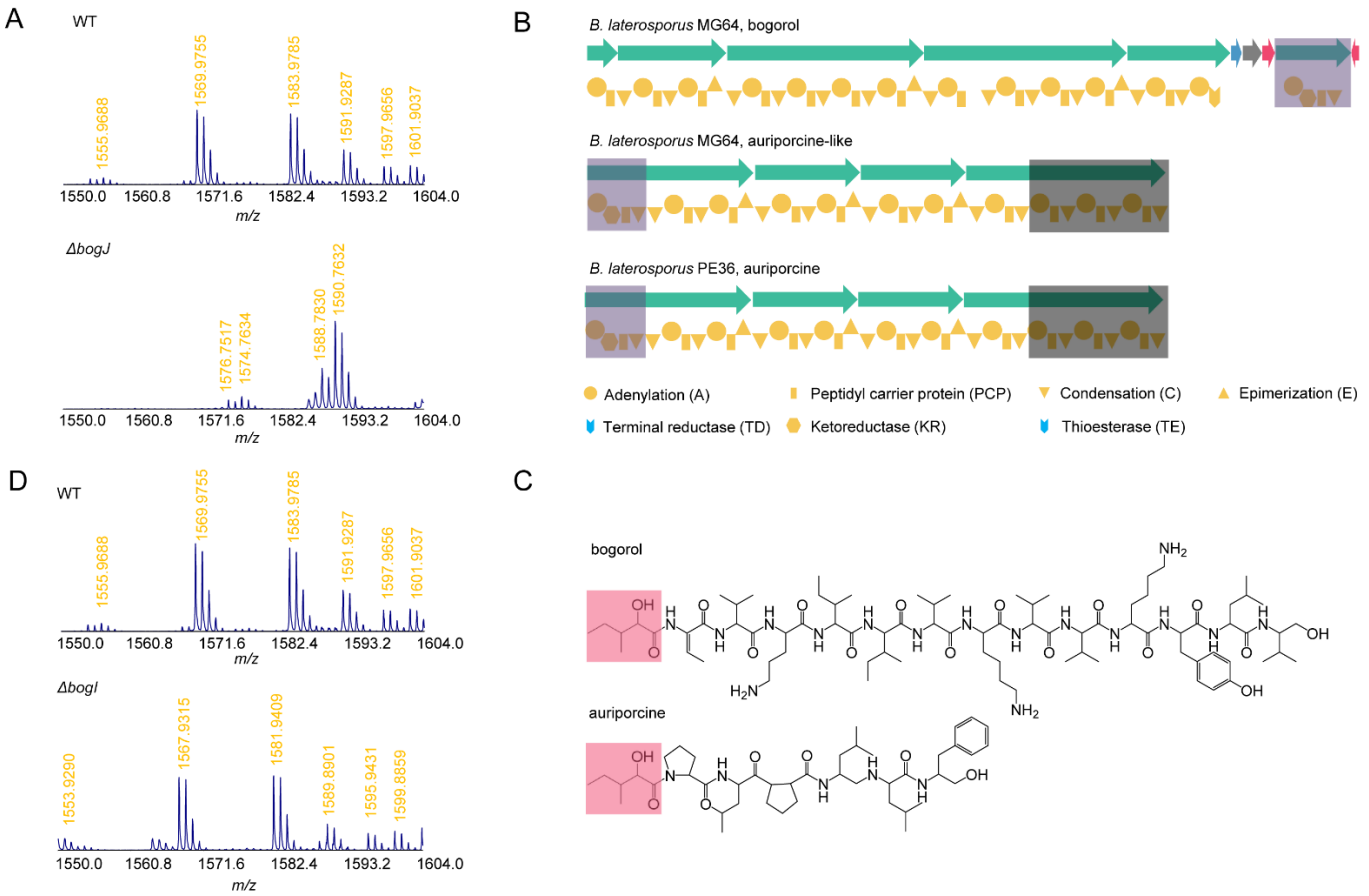
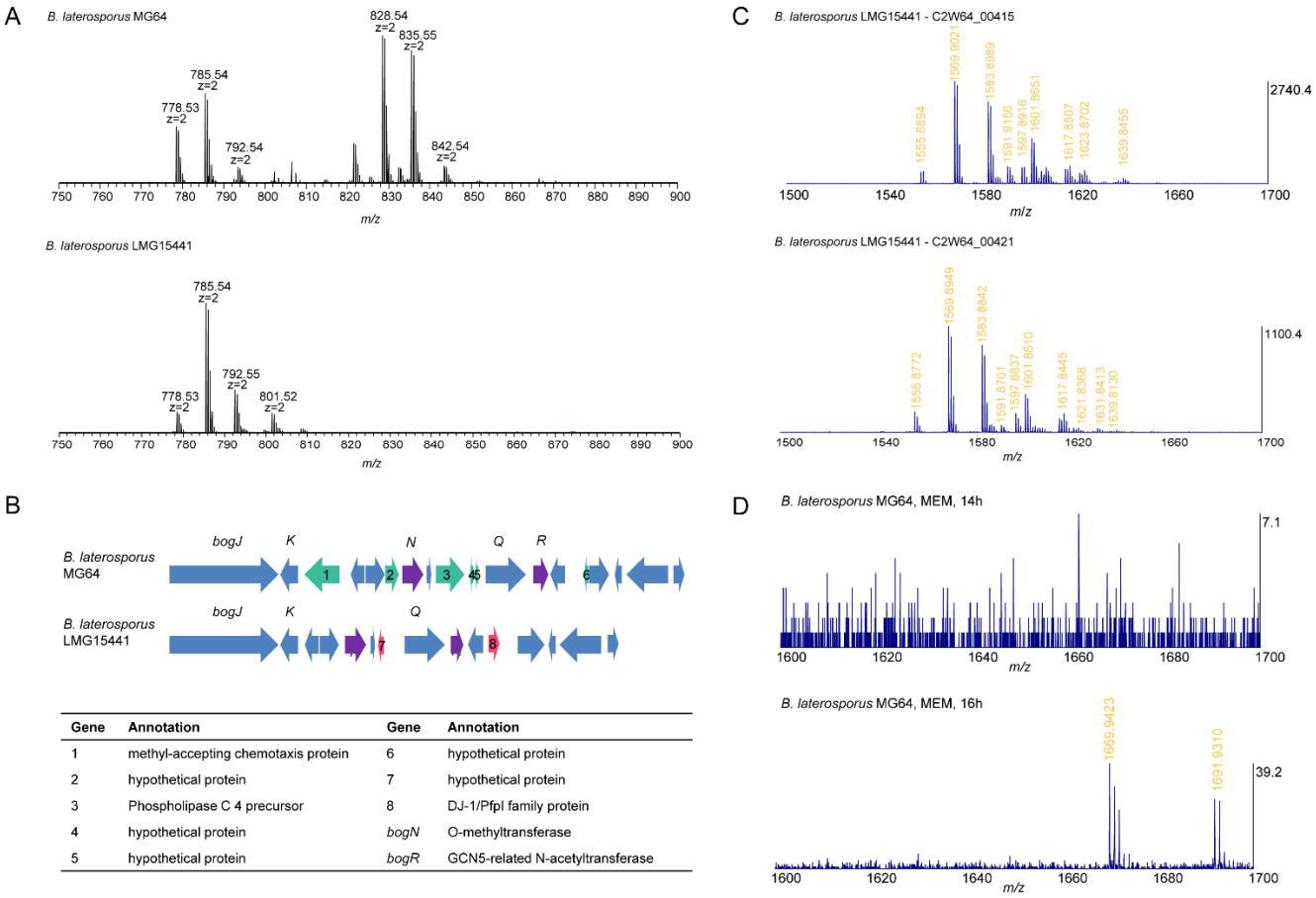


Fig. S4. Investigation of lipoinitiation and valinol formation of bogorols. (A) MALDI-TOF analysis of extracts from wild type (WT) and *ΔbogJ* mutant of *B. laterosporus* LMG15441. The $[M + H]^+$ of representative compounds are indicated. (B) Biosynthetic gene cluster of bogorol and auriporcine. The modules responsible for fatty acid incorporation are indicated with purple shading. The unfunctional modules in auriporcine BGC are indicated in grey shading. (C) The structures of bogorol and auriporcine. The fatty acids of both compounds are indicated with red shading. (D) MALDI-TOF analysis of extracts from wild type (WT) and *ΔbogI* mutant of *B. laterosporus* LMG15441. The $[M + H]^+$ of representative compounds were indicated.



Gene	Annotation	Gene	Annotation
1	methyl-accepting chemotaxis protein	6	hypothetical protein
2	hypothetical protein	7	hypothetical protein
3	Phospholipase C 4 precursor	8	DJ-1/Pfpl family protein
4	hypothetical protein	*bogN*	O-methyltransferase
5	hypothetical protein	*bogR*	GCN5-related N-acetyltransferase

Fig. S5. Investigation of succinylation of succilins. (A) LC-MS of extracts from *B. laterosporus* MG64 and LMG15441.z in the labels indicate the charges of the ions. (B) Flanking genes of the bogorol BGC in *B. laterosporus* MG64 and LMG15441. Blue, present in both strains; green, only present in *B. laterosporus* MG64; red, only present in *B. laterosporus* LMG15441; purple, transferase. The table presents the annotation of some of the genes. (C) MALDI-TOF analysis of extracts from *B. laterosporus* LMG15441 with heterologous expression of the gene *bogN* (C2W64_00415, from *B. laterosporus* MG64) and *bogR* (C2W64_00421, from *B. laterosporus* MG64). (D) MALDI-TOF analysis of extracts from *B. laterosporus* MG64 at different time points. MEM is the medium used in for this experiment.

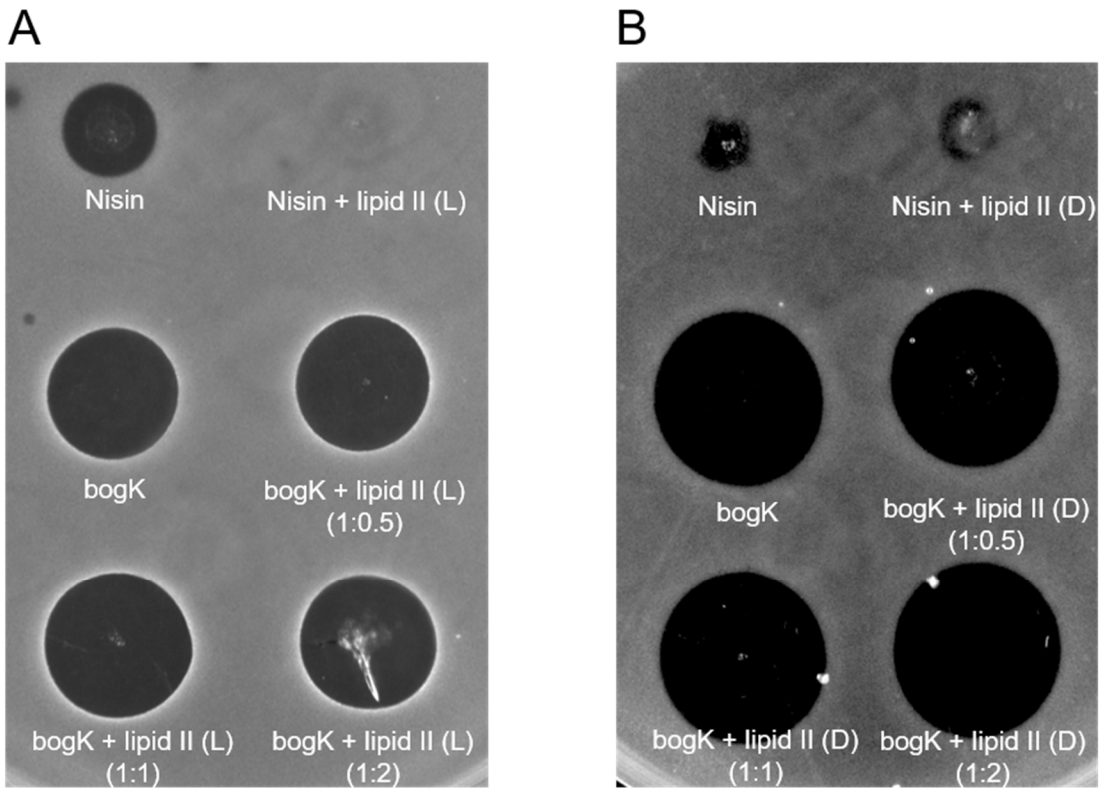


Fig. S6. Investigation of Lipid II binding property of bogorol K. (A) Investigation of bogorol K binding to Gram-positive type Lipid II. *Staphylococcus aureus* is used as an indicator. (B) Investigation of bogorol K binding to Gram-negative type Lipid II. *Xanthomonas campestris* is used as an indicator. Lipid II was mixed with compounds at different ratios before spotting onto the plate, which contains an indicator strain. Nisin was used as a control. L or D in the bracket means a Gram-positive or Gram-negative type of lipid II was used. The plate was incubated at 28 °C overnight before recording the result.

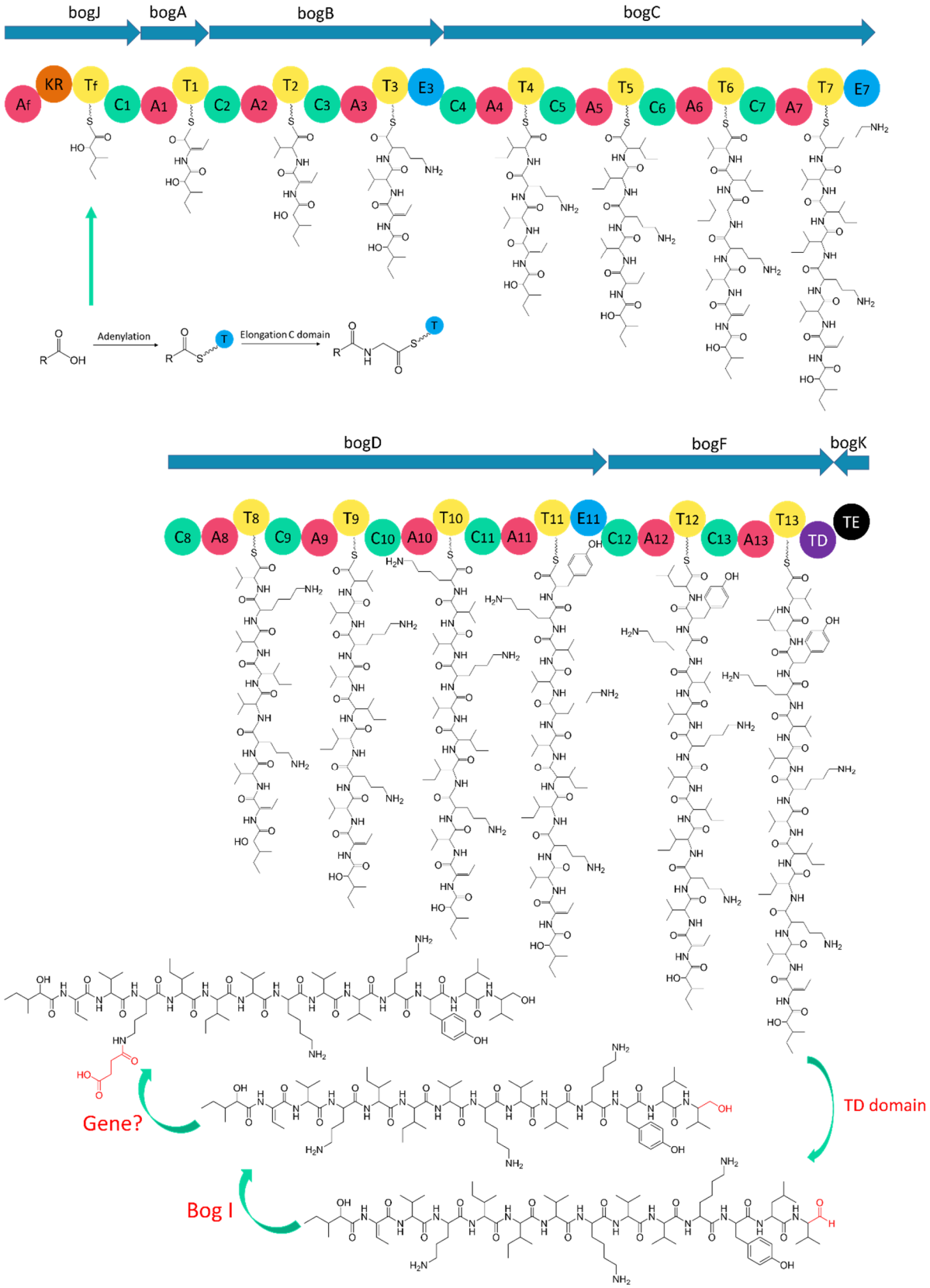


Fig. S7. Summary of the biosynthetic pathway of bogorols and succilins. A, adenylation domain; KR, ketoreductase domain; T, thiolation domain; C, condensation domain; E, epimerization domain; TD, terminal reductase domain; TE, thioesterase domain.

Table S1. Partial chemical shift (ppm) assignments of bogorol K (d_6-DMSO).

Residue	NH	^{1}Hα	^{1}Hβ	Others
FA		3.79	$OH^{\beta1}$, 5.61; $CH^{\beta2}$, 1.71	$CH_2^{\gamma1}$, 1.38/1.12; $CH_3^{\gamma2}$, 0.88; $CH_3^{\delta1}$, 0.80
Dhb-1	9.15		5.93	$CH_3^{\gamma1}$, 1.78
Val-2	7.81	4.24	2.11	CH_3^{γ}, 0.87
Orn-3	8.08	4.42	1.68	CH_2^{γ}, 1.55; CH_2^{δ}, 2.76; NH_2^{ε}, 7.63
Ile-4	7.85	4.28	1.70	$CH_2^{\gamma1}$, 1.35/1.02*
Ile-5	8.00	4.17	1.69	$CH_2^{\gamma1}$,1.43/1.04
Val-6	7.81	4.18	1.93	
Lys-7	7.93	4.38	1.63	CH_2^{γ}, 1.26; CH_2^{δ}, 1.48; CH_2^{ε}, 2.70; NH_2^{ζ},7.64
Val-8	7.86	4.22	1.91	
Val-9	7.88	4.11	1.94	
Lys-10	7.78	4.24	1.43/1.32*	CH_2^{γ}, 1.05; CH_2^{δ}, 1.42; CH_2^{ε}, 2.65; NH_2^{ζ}, 7.65
Tyr-11	8.09	4.47	2.84/2.60	CH^{δ}, 6.99; CH^{ε}, 6.62
Leu-12	8.08	4.24	1.38	$CH_3^{\delta1}$, 0.82; $CH_3^{\delta2}$, 0.76*
Valinol-13	7.40	3.53	$CH_2^{\beta1}$, 3.34; $CH^{\beta2}$, 1.80	CH3γ, 0.81

*Tentative assignment due to overlap, but in accordance with the literature (1).

Table S2. Partial chemical shift (ppm) assignments of bogorol L (d_6-DMSO).

Residue	NH	^{1}Hα	^{1}Hβ	Others
FA		3.79	$OH^{\beta1}$, 5.61*; $CH^{\beta2}$, 1.72	$CH_2^{\gamma1}$, 1.38/1.13; $CH_3^{\gamma2}$, 0.88; $CH_3^{\delta1}$, 0.80
Dhb-1	9.11		5.97	$CH_3^{\gamma1}$, 1.79
Val-2	7.79	4.24	2.11	CH_3^{γ}, 0.87
Lys-3	8.08	4.34	1.63/1.55	CH_2^{γ}, 1.28; CH_2^{δ}, 1.48; CH_2^{ε}, 2.70;
Ile-4	7.79	4.24	1.70	
Ile-5	7.99	4.17	1.71	$CH_2^{\gamma1}$,1.41/1.05
Val-6	7.78	4.17	1.94	
Lys-7	7.94	4.37	1.63	CH_2^{γ}, 1.27; CH_2^{δ}, 1.48; CH_2^{ε}, 2.70;
Val-8	7.86	4.21	1.93	
Val-9	7.88	4.11	1.94	
Lys-10	7.78	4.24	1.42/1.31*	CH_2^{γ}, 1.05; CH_2^{δ}, 1.42; CH_2^{ε}, 2.65;
Tyr-11	8.09	4.47	2.84/2.61	CH^{δ}, 6.99; CH^{ε}, 6.62
Leu-12	8.09	4.24	1.39	CH^{γ}, 1.35*; $CH_3^{\delta1}$, 0.81; $CH_3^{\delta2}$, 0.76
Valinol-13	7.40	3.54	$CH_2^{\beta1}$, 3.34; $CH^{\beta2}$, 1.81	CH_3^{γ}, 0.81

*Tentative assignment due to overlap, but in accordance with the literature (1).

Table S3. Marfey analysis of bogorol K (**3**) and bogorol L (**4**).

Standard	**Retention Time (min.)**	**bogorol K (3)**	**bogorol L (4)**
L-Val	10.30	10.25	10.21
D-Val	12.05	-	-
L-Leu	12.06	12.02	11.98
D-Leu	13.76	-	-
L-Ile	11.76	11.71	11.68
D-Ile	13.60	-	-
L-Phe (standard)	11.98	11.93	11.89
L-Tyr	6.59 / 9.14	-	-
D-Tyr	6.58 / 9.77	6.44 / 9.69	6.41 / 9.65
L-Orn	4.55 / 4.91	-	-
D-Orn	4.04 / 4.89	3.80 / 4.69	-
L-Lys	5.14 / 5.61	4.84 / 5.39	4.85 / 5.38
D-Lys	5.12 / 4.78	4.78 / 4.50	4.79 / 4.51
L-(S)Vol	9.94	9.87	9.84
D-(R)Vol	12.05	-	-

Table S4. Partial chemical shift (ppm) assignments of succilin K (d_6-DMSO).

Residue	NH	^{1}Hα	^{1}Hβ	Others
FA		3.77	$OH^{\beta1}$, 5.61; $CH^{\beta2}$, 1.72	$CH_2^{\gamma1}$, 1.37/1.12; $CH_3^{\gamma2}$, 0.88; $CH_3^{\delta1}$, 0.79
Dhb-1	9.08		5.99	$CH_3^{\gamma1}$, 1.79
Val-2	7.76	4.25	2.10	CH_3^{γ}, 0.86
Orn-3 (Suc)	8.12	4.31	1.63/1.52	CH_2^{γ}, 1.41/1.33; CH_2^{δ}, 2.98; NH^{ε}, 7.79; **CH_2^{suc1}, 2.28; CH_2^{suc2}, 2.36**
Ile-4	7.78	4.21	1.71	
Ile-5	8.01	4.16	1.72	$CH_2^{\gamma1}$,1.43/1.05
Val-6	7.84	4.16	1.94	
Lys-7	8.01	4.35	1.63	CH_2^{γ}, 1.27; CH_2^{δ}, 1.48; CH_2^{ε}, 2.70;
Val-8	7.89	4.20	1.93	
Val-9	7.89	4.10	1.94	
Lys-10	7.82	4.24	1.41/1.32*	CH_2^{γ}, 1.04; CH_2^{δ}, 1.41; CH_2^{ε}, 2.65;
Tyr-11	8.11	4.47	2.84/2.61	CH^{δ}, 6.99; CH^{ε}, 6.62
Leu-12	8.10	4.24	1.39	CH^{γ}, 1.35*; $CH_3^{\delta1}$, 0.82; $CH_3^{\delta2}$, 0.76
Valinol-13	7.40	3.53	$CH_2^{\beta1}$, 3.34; $CH^{\beta2}$, 1.80	$CH_3^{\gamma1}$, 0.82; $CH_3^{\gamma2}$, 0.80

*Tentative assignment due to overlap, but in accordance with the literature (1).

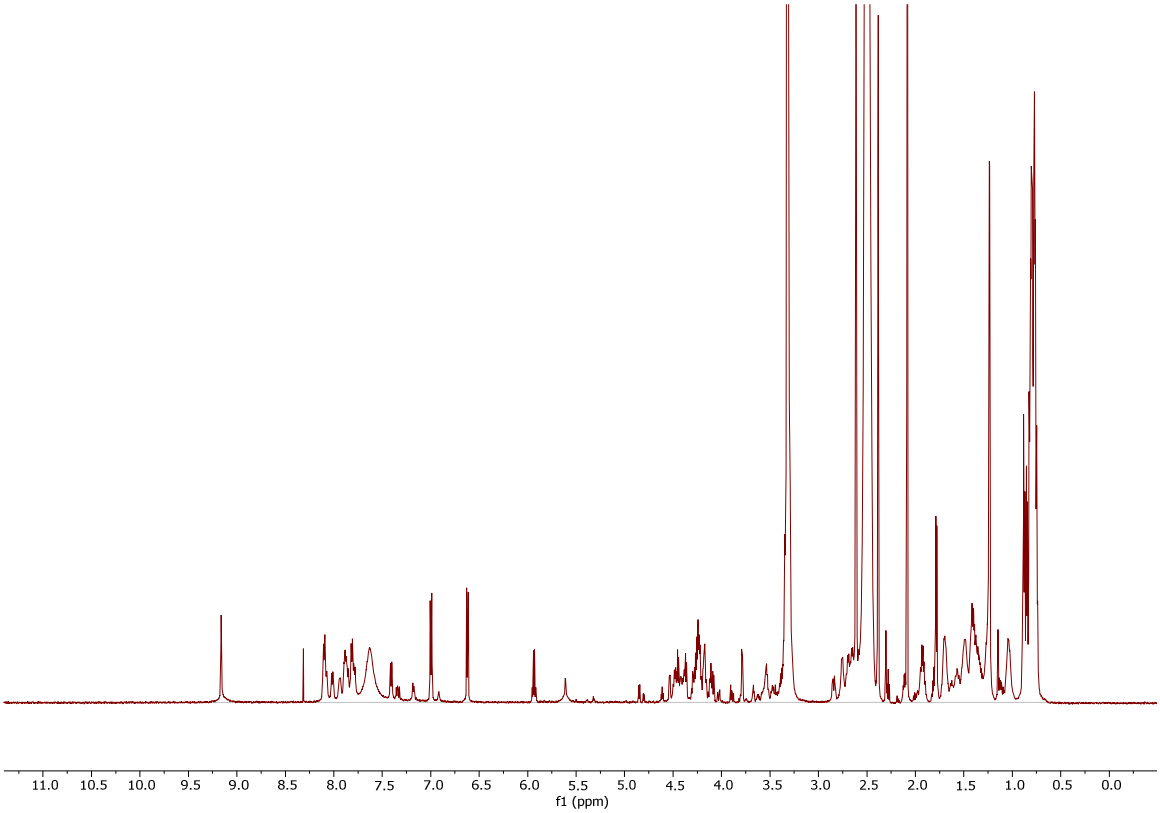
Table S5. Comparison of bogorol and succilin peptides

Position	FA	1	2	3	4	5	6	7	8	9	10	11	12	13
Bogorol A	$C_6H_{11}O_2$	Dhb	Leu	Orn	Ile	Val	Val	Lys	Val	Leu	Lys	Tyr	Leu	Valinol
Bogorol B	$C_6H_{11}O_2$	Dhb	Val	Orn	Ile	Val	Val	Lys	Val	Leu	Lys	Tyr	Leu	Valinol
Bogorol C	$C_6H_{11}O_2$	Dhb	Val	Orn	Val	Val	Val	Lys	Val	Leu	Lys	Tyr	Leu	Valinol
Bogorol D	$C_6H_{11}O_2$	Dhb	Met	Orn	Ile	Val	Val	Lys	Val	Leu	Lys	Tyr	Leu	Valinol
Bogorol E	$C_6H_{11}O_2$	Dhb	Met-O	Orn	Ile	Val	Val	Lys	Val	Leu	Lys	Tyr	Leu	Valinol
Brevibacillin	$C_6H_{11}O_2$	Dhb	Leu	Orn	Ile	Ile	Val	Lys	Val	Val	Lys	Tyr	Leu	Valinol
Brevibacillin V	$C_6H_{11}O_2$	Dhb	Leu	Orn	Ile	Val	Val	Lys	Val	Val	Lys	Tyr	Leu	Valinol
BT peptide	$C_{11}H_{20}O_2N_1$	Dhb	Leu	Orn	Ile	Val	Val	Lys	Val	Leu	Lys	Tyr	Leu	Valinol
Bogorol I (**1**)	$C_6H_{11}O_2$	Dhb	Val	Orn	Ile	Val	Val	Lys	Val	Val	Lys	Tyr	Leu	Valinol
Bogorol J (**2**)	$C_6H_{11}O_2$	Dhb	Val	Orn	Val	Ile	Val	Lys	Val	Val	Lys	Tyr	Leu	Valinol
Bogorol K (**3**)	$C_6H_{11}O_2$	Dhb	Val	Orn	Ile	Ile	Val	Lys	Val	Val	Lys	Tyr	Leu	Valinol
Bogorol L (**4**)	$C_6H_{11}O_2$	Dhb	Val	Lys	Ile	Ile	Val	Lys	Val	Val	Lys	Tyr	Leu	Valinol
Succilin I (**5**)	$C_6H_{11}O_2$	Dhb	Val	Orn-S	Ile	Val	Val	Lys	Val	Val	Lys	Tyr	Leu	Valinol
Succilin J (**6**)	$C_6H_{11}O_2$	Dhb	Val	Orn-S	Val	Ile	Val	Lys	Val	Val	Lys	Tyr	Leu	Valinol
Succilin K (**7**)	$C_6H_{11}O_2$	Dhb	Val	Orn-S	Ile	Ile	Val	Lys	Val	Val	Lys	Tyr	Leu	Valinol
Succilin L (**8**)	$C_6H_{11}O_2$	Dhb	Val	Lys-S	Ile	Ile	Val	Lys	Val	Val	Lys	Tyr	Leu	Valinol

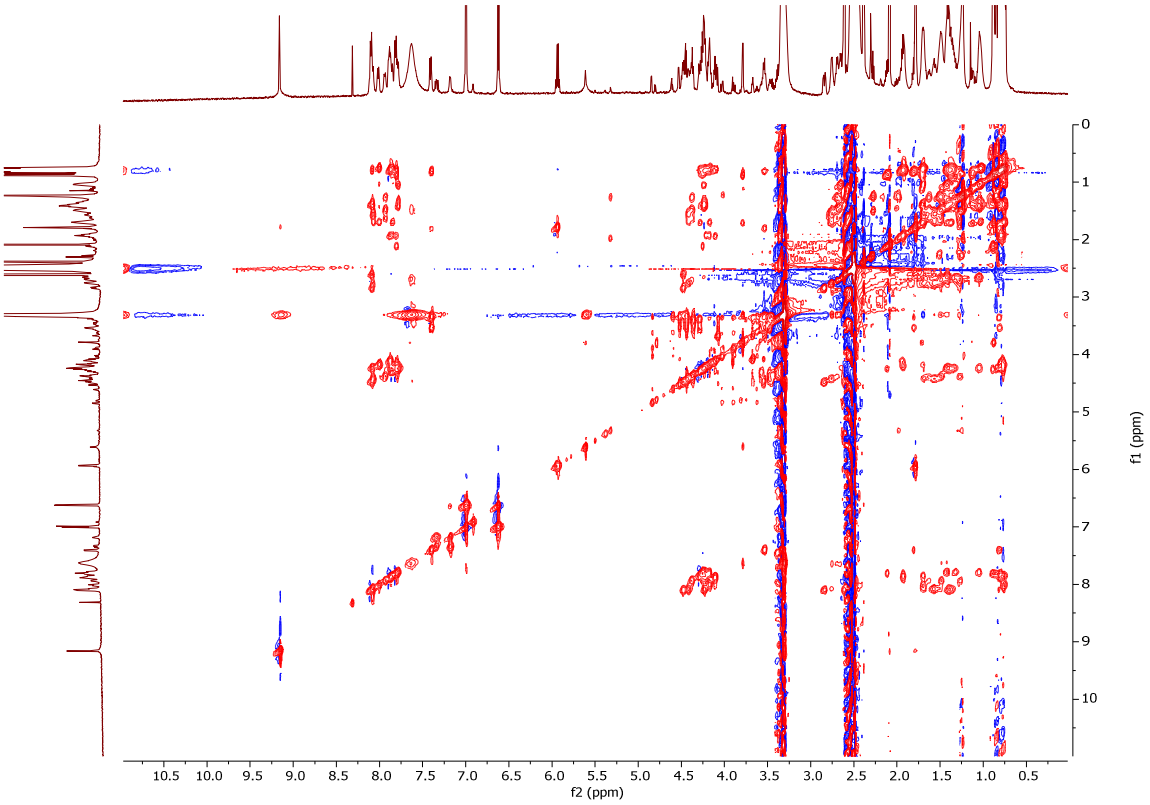
Note: The position number of conserved residues are indicated in black. The newly identified peptides and their closest peptides are indicated in the same background color, and the different residues in the characterized peptides are indicated in red font. FA: fatty acid.

Original NMR spectra

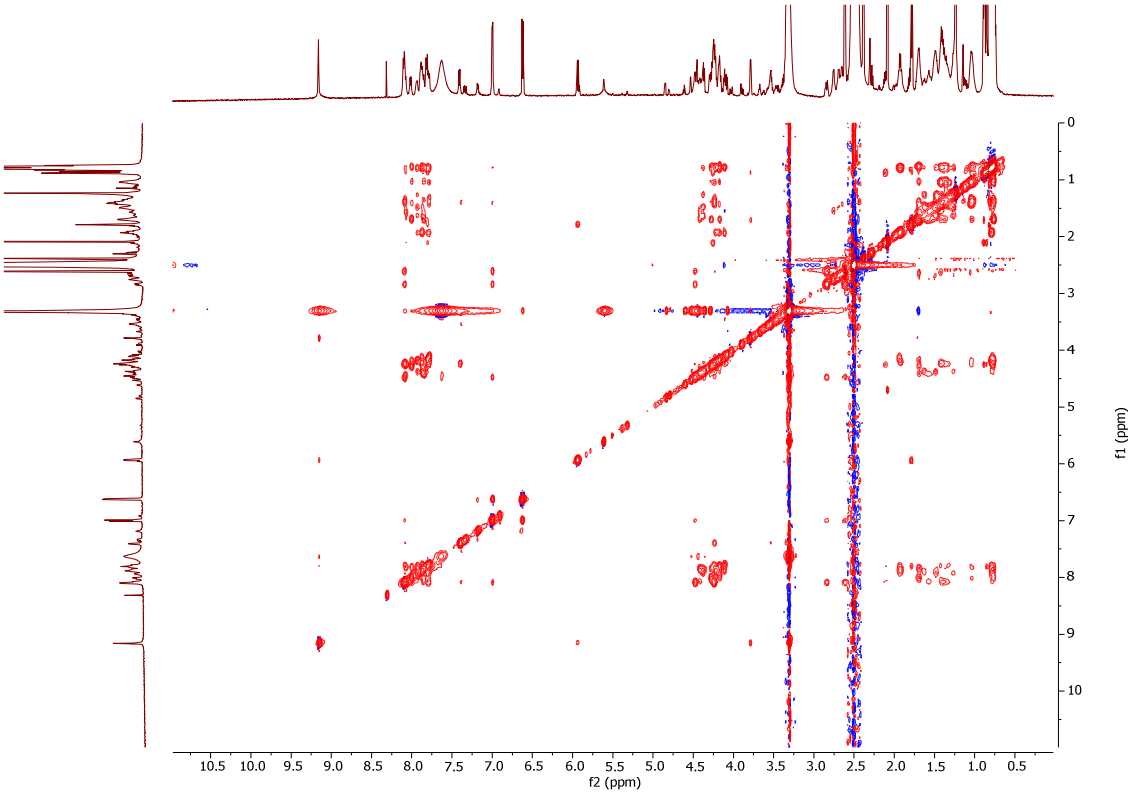
^{1}H NMR of bogorol K (**3**)



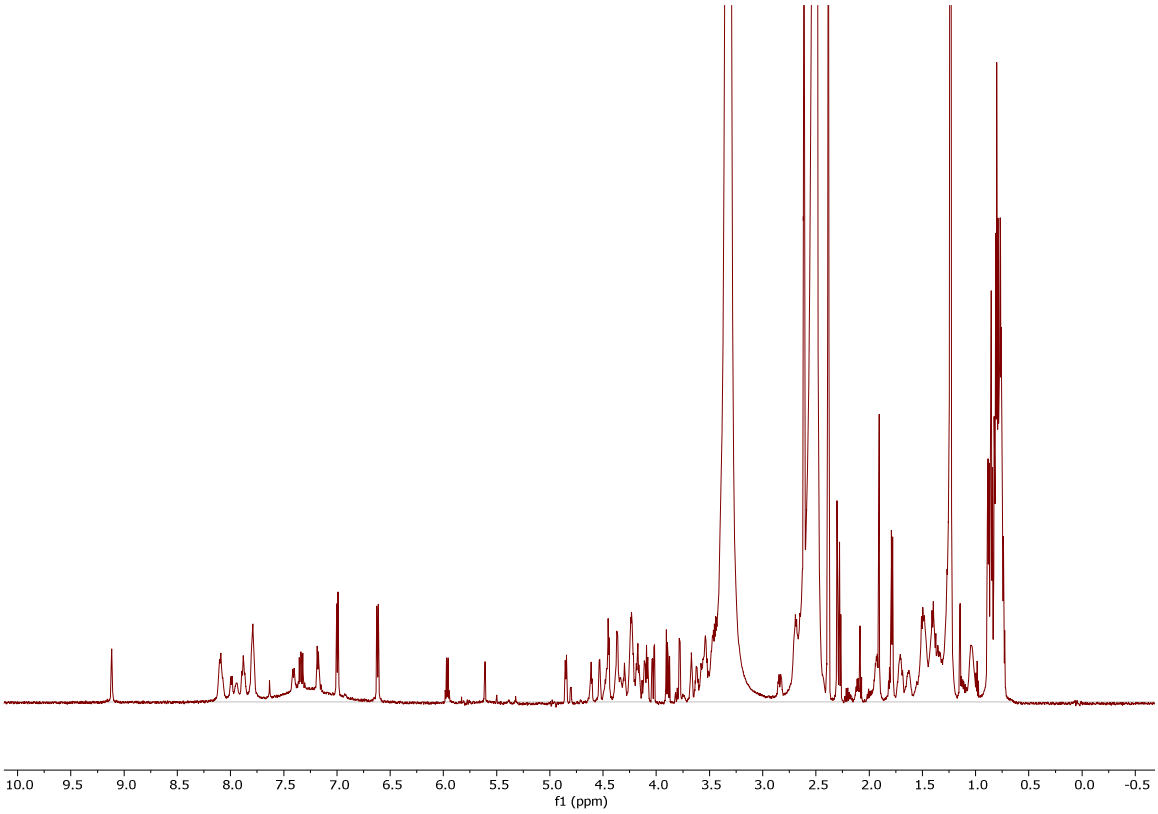
^{1}H-^{1}H-TOCSY NMR of bogorol K (**3**)



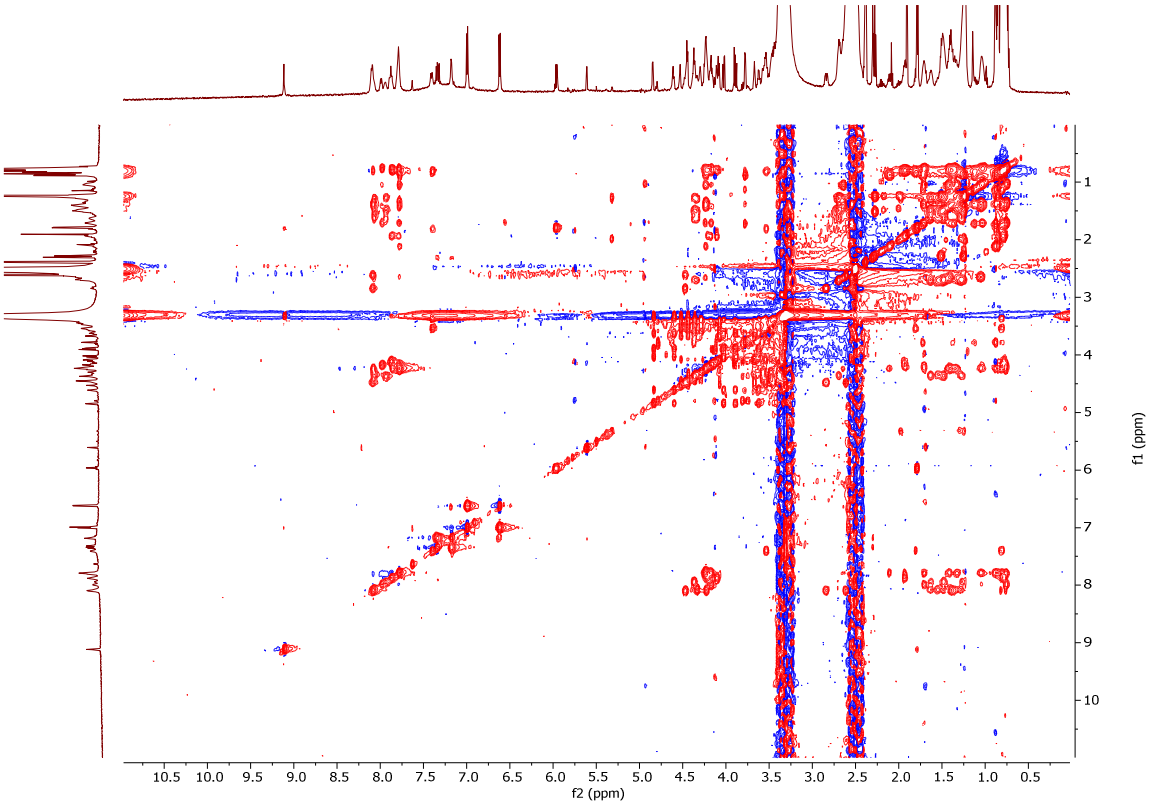
^{1}H-^{1}H-NOESY NMR of bogorol K (**3**)



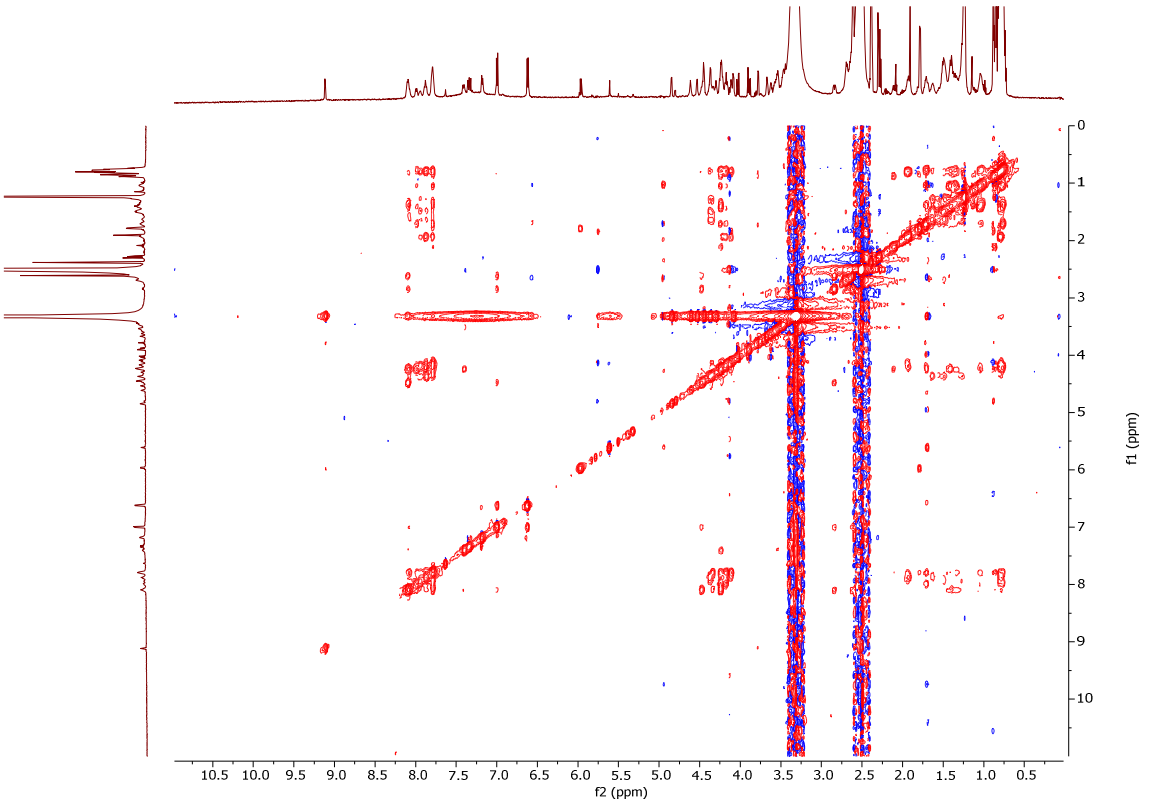
^{1}H NMR of bogorol L (**4**)



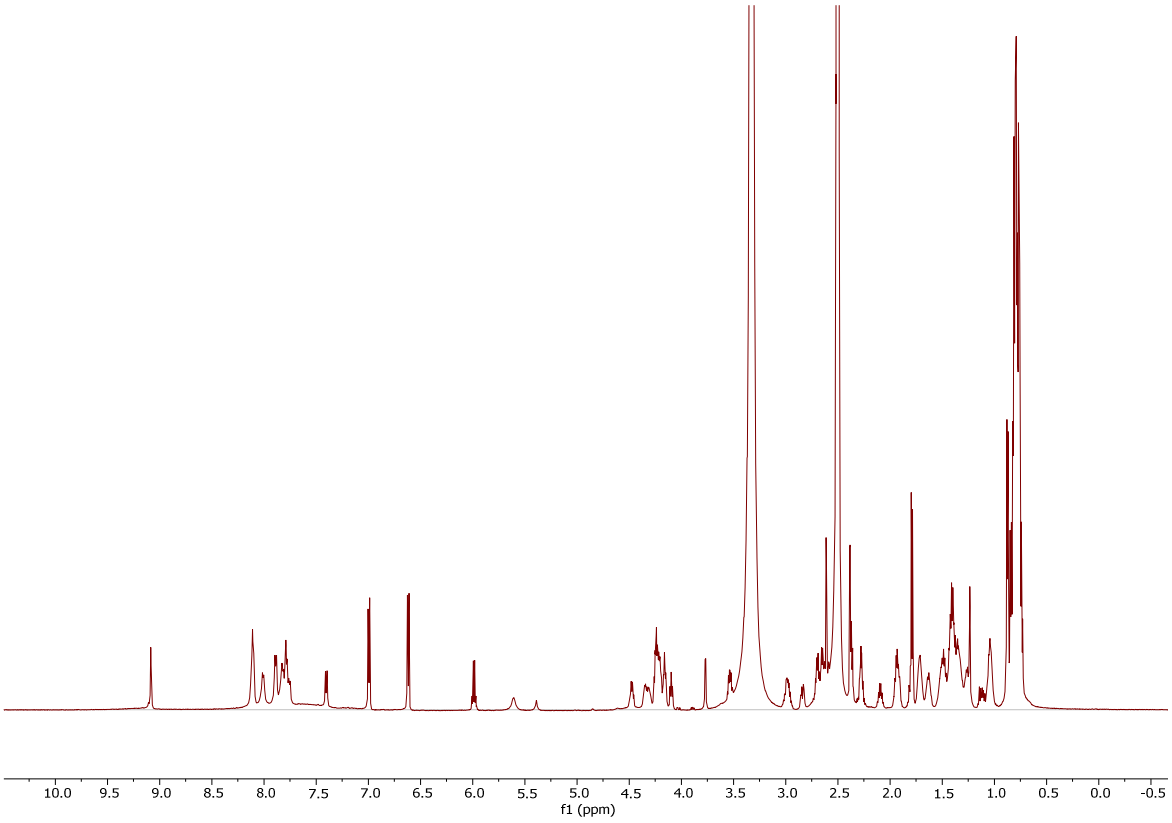
^{1}H-^{1}H-TOCSY NMR of bogorol L (**4**)



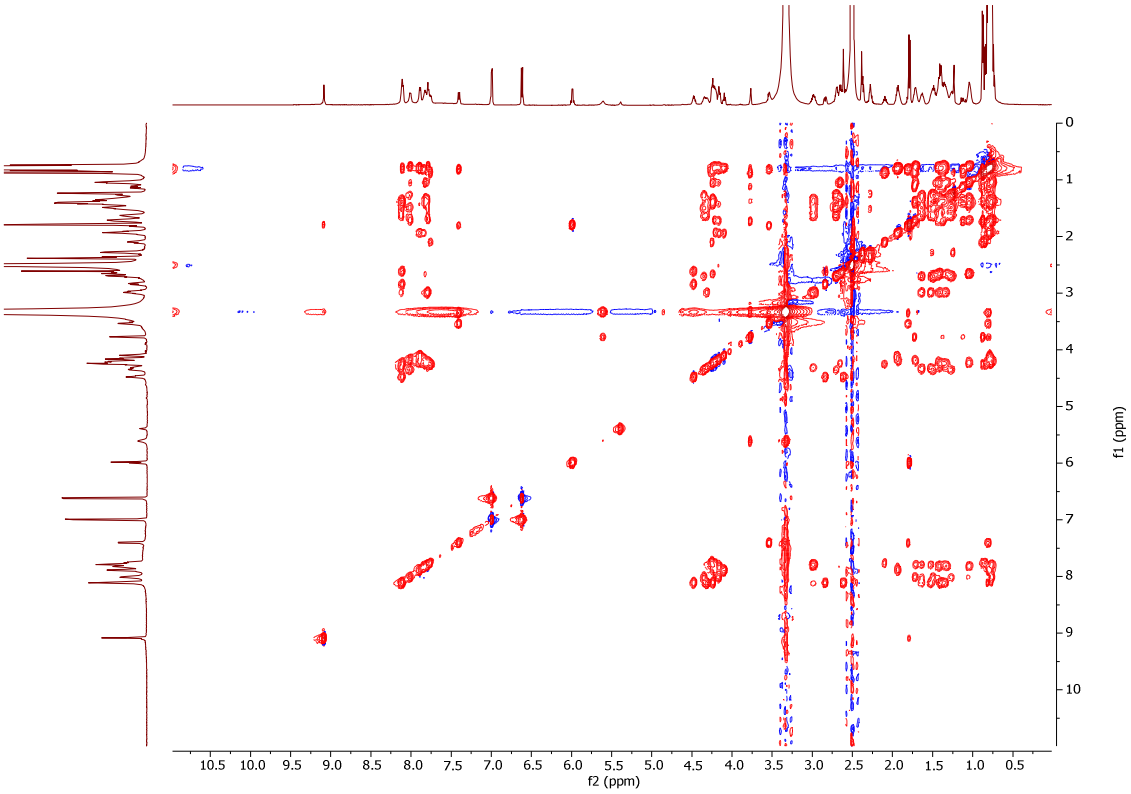
^{1}H-^{1}H-NOESY NMR of bogorol L (**4**)



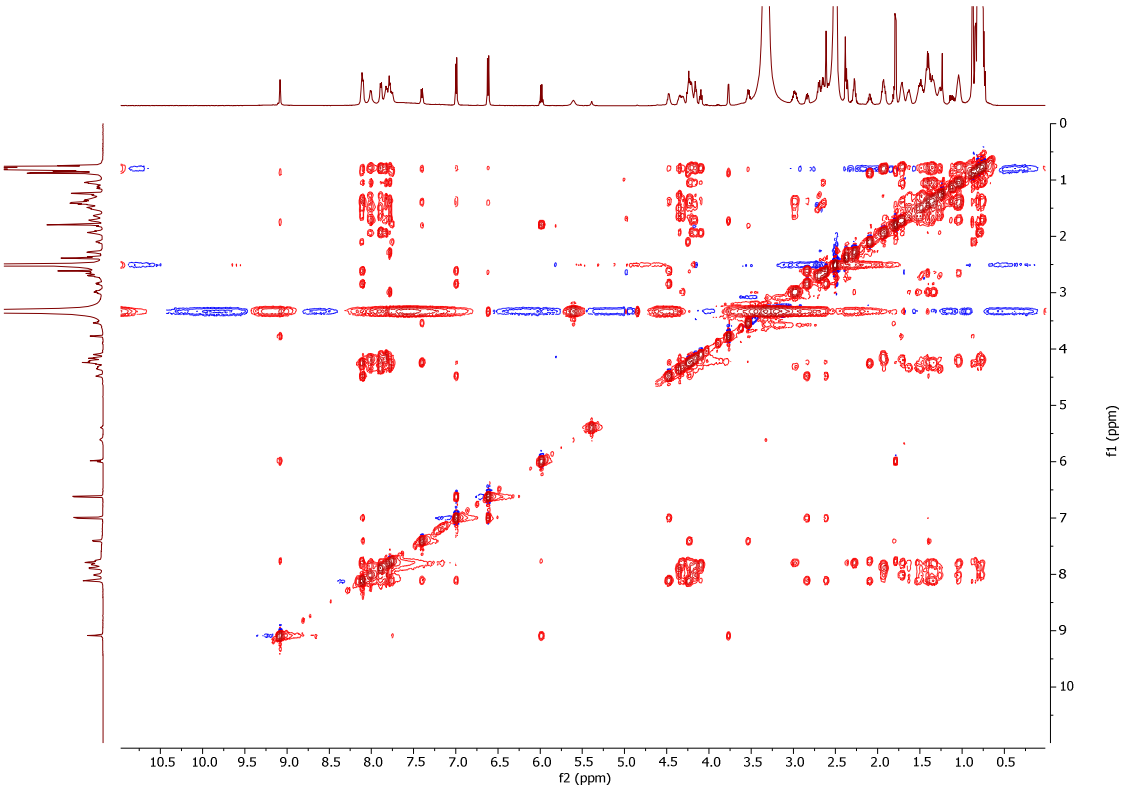
^{1}H NMR of succilin K (**7**)



^{1}H-^{1}H-TOCSY NMR of succilin K (**7**)



^{1}H-^{1}H-NOESY NMR of succilin K (**7**)



Reference

1. **Yang X, Huang E, Yuan C, Zhang L, Yousef AE.** 2016. Isolation and structural elucidation of brevibacillin, an antimicrobial lipopeptide from *Brevibacillus laterosporus* that combats drug-resistant Gram-positive bacteria. Appl Environ Microbiol **82:**2763-2772.