Electronic Supplementary Material (ESI) for Natural Product Reports.

The Phosphate Ester Group in Secondary Metabolites

Supporting Information

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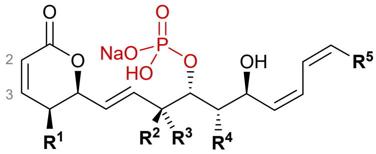
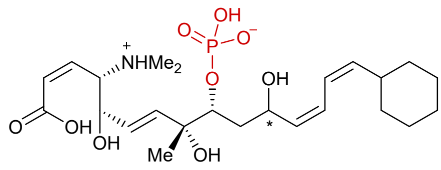
Figure S1. Polyketides

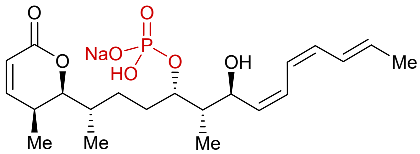
Fostriecin Family



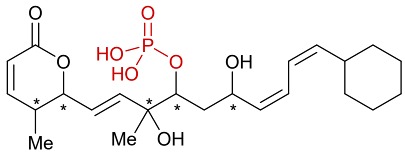
	R^1	R^2	R^3	R^4	R^5
Fostriecin (**1**)	H	Me	OH	H	(*E*)-CH=CHCH$_2$OH
PD 113,270 (**2**)	H	Me	OH	H	(*E*)-CH=CHCH$_3$
PD 113,271 (**3**)	OH	Me	OH	H	(*E*)-CH=CHCH$_2$OH
Phostriecin (**4**)	OH	H	H	Me	(*E*)-CH=CH(CH$_2$)$_4$CH$_3$
Phosdiecin A (**5**)	OH	H	H	Me	(CH$_2$)$_2$CH(Me)$_2$
Phosdiecin B (**6**)	OH	H	H	Me	(CH$_2$)$_2$CH(Me)$_2$, (saturated at C2-C3)



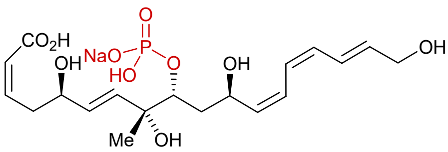
Phoslactomycin H (**18**)



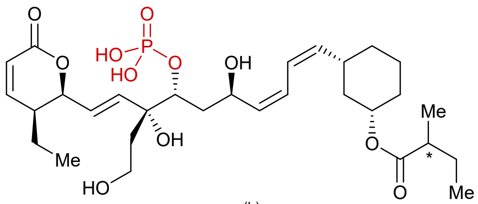
Cytostatin (**7**)



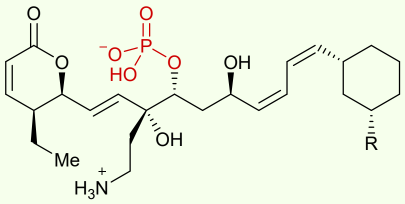
Phoslactomicyn I (**19**)



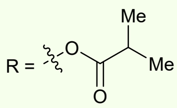
PD 114,027 (**S1**)[(a)]



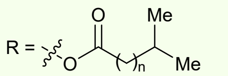
S2[(b)]



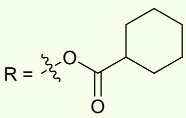
Phoslactomycin B (**9**) R = H
Phoslactomycin G/
leustroducsin H (**14**) R = OH



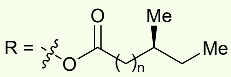
Phoslactomicyn A (**8**)



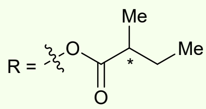
Phoslactomicyn C/
phosphazomycin C$_1$ (**10**) n = 1
Phoslactomicyn D (**11**) n = 2
Leustroducsin A (**15**) n = 3
S4 n = 4
Leustroducsin C (**17**) n = 5



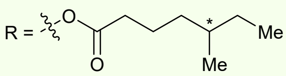
Phoslactomicyn E (**12**)



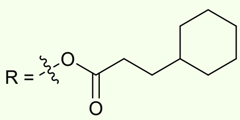
Phoslactomycin F (**13**) n = 2
Leustroducsin B (**16**) n = 4



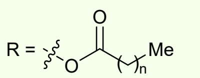
Phosphazomycin C$_2$ (**S3**)



Phoslactomycin I–h (**S5**)



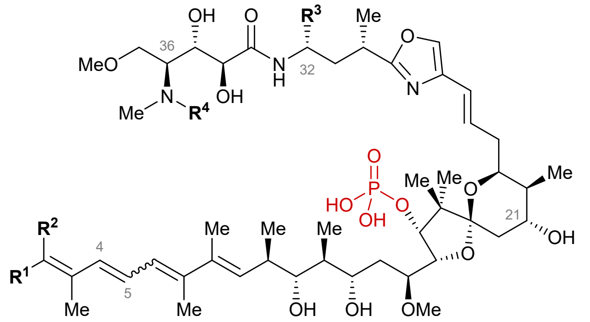
Phoslactomycin I–i (**S6**)



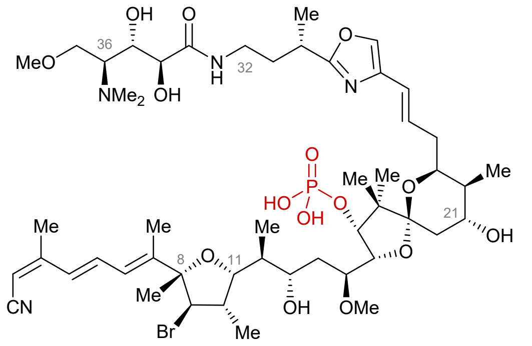
Phoslactomycin I–j (**S7**) n = 6
S8 n = 2

(a) Stereochemistry according to: D. S. Lewy, C.-M. Gauss, D. R. Soenen and D. L. Boger, *Curr. Med. Chem.*, 2002, **9**, 2005–2032; (b) Stereochemistry according to: B. M. Trost, J. D. Knopf and C. S. Brindle, *Chem. Rev.*, 2016, **116**, 15035–15088.

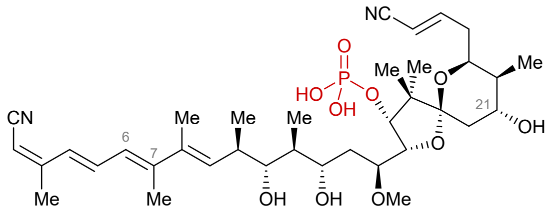
Calyculin Family



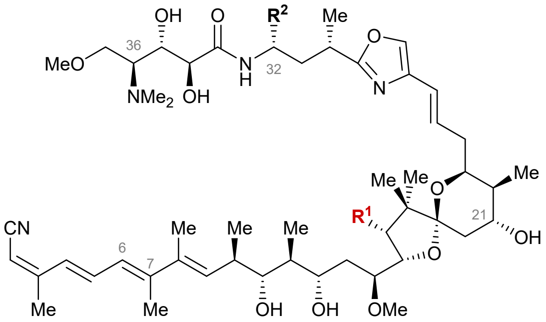
	C4=C5	R^1	R^2	R^3	R^4
Calyculin A (**23**)	*E*	H	CN	H	Me
Calyculin B (**24**)	*E*	CN	H	H	Me
Calyculin C (**25**)	*E*	H	CN	Me	Me
Calyculin D (**26**)	*E*	CN	H	Me	Me
Calyculin E (**27**)	*Z*	H	CN	H	Me
Calyculin F (**28**)	*Z*	CN	H	H	Me
Calyculin G (**29**)	*Z*	H	CN	Me	Me
Calyculin H (**30**)	*Z*	CN	H	Me	Me
des-*N*-Me Calyculin A (**31**)	*E*	H	CN	H	H
Calyculinamide A (**32**)	*E*	H	$CONH_2$	H	Me
Calyculinamide B (**33**)	*E*	$CONH_2$	H	H	Me
Calyculinamide F (**34**)	*Z*	H	$CONH_2$	H	Me



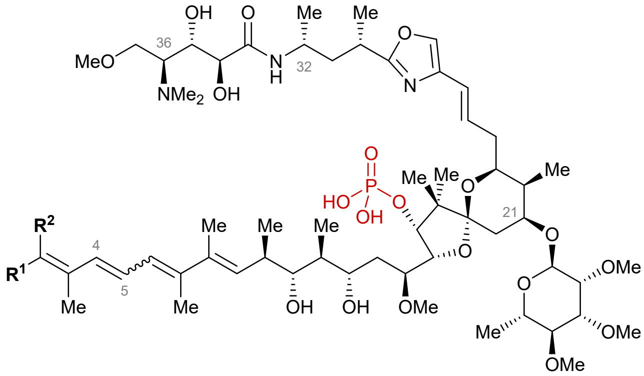
Calyculin J (**38**)



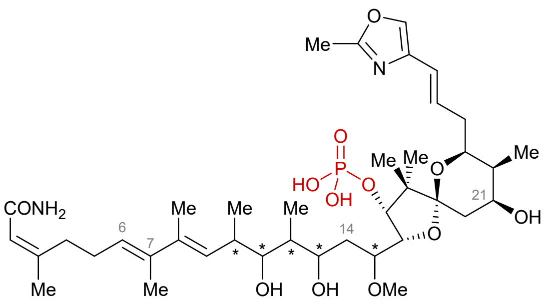
Hemicalyculin A (**39**)



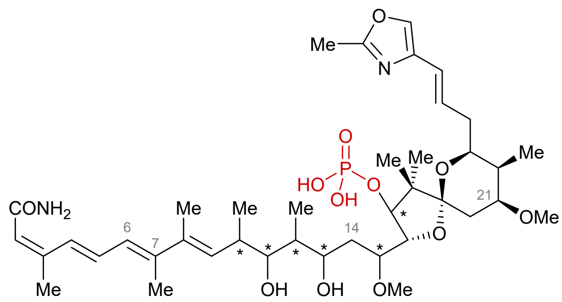
	R^1	R^2
Dephosphonocalyculin A (**35**)	OH	H
Phosphocalyculin A (**36**)	$OP_2O_6H_3$	H
Phosphocalyculin C (**37**)	$OP_2O_6H_3$	Me



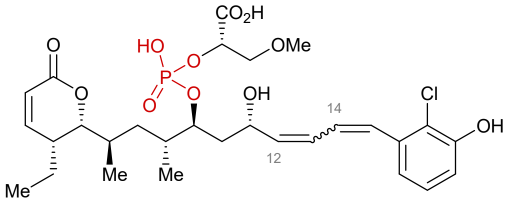
	C4=C5	R^1	R^2
Clavosine A (**40**)	*E*	H	$CONH_2$
Clavosine B (**41**)	*E*	$CONH_2$	H
Clavosine C (**42**)	*Z*	$CONH_2$	H



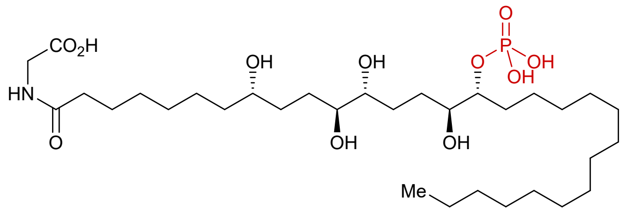
Geometricin A (**43**)



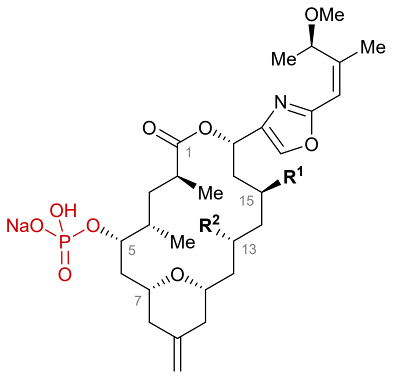
Swinhoeiamide A (**44**)



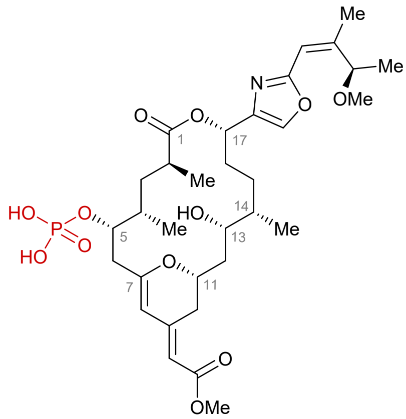
Franklinolide A (**20**)	12Z, 14Z
Franklinolide B (**21**)	12E, 14E
Franklinolide C (**22**)	12E, 14Z



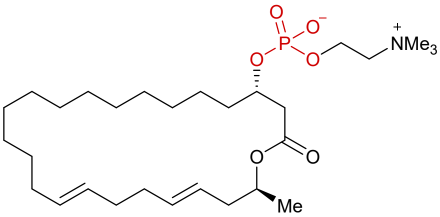
Phosphoeleganin (**45**)



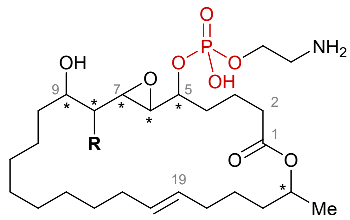
	R^1	R^2
Enigmazole A (**46**)	OH	H
15-*O*-Methylenigmazole A (**47**)	OMe	H
13-*OH*-15-*O*-Methylenigmazole A (**48**)	OMe	OH



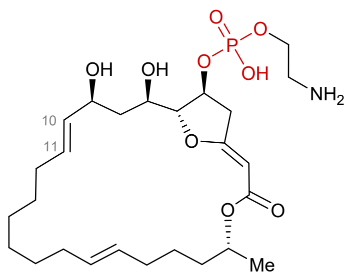
Proposed structure of
(–)-Enigmazole B (**S9**)



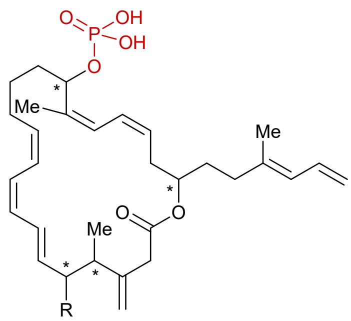
Eushearilide (**49**)



	R	H6-H7
JBIR-19 (**50**)	OH	*trans*
JBIR-20 (**51**)	H	*cis*



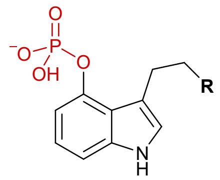
Preussolide A (**52**) (saturated at C10-C11)
Preussolide B (**53**)



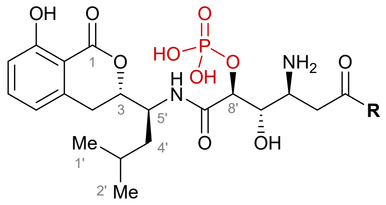
	R
Difficidin (**S10**)	H
Oxydifficidin (**S11**)	OH

*Absolute configuration not determined.

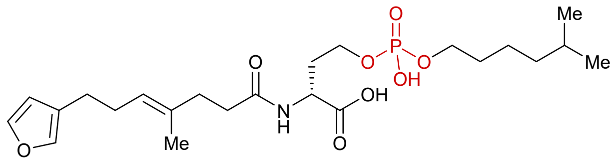
Figure S2. Alkaloids and Related Secondary Metabolites



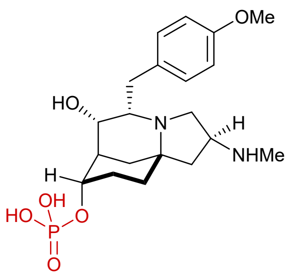
	R
Psilocybin (**54**)	$NHMe_2^+$
Baeocystin (**57**)	NH_2Me^+
Norbaeocystin (**58**)	NH_3^+
Aeruginascin (**59**)	NMe_3^+



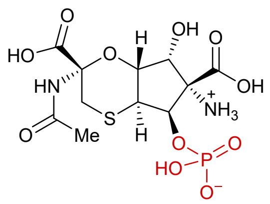
	R
Amicoumacin A 8'-phosphate (**60**)	NH_2
Amicoumacin B 8'-phosphate (**61**)	OH



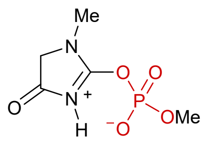
Pokepola ester (**S12**)



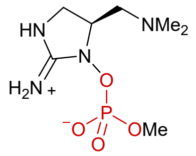
FR901483 (**64**)



Tagetitoxin (**65**)

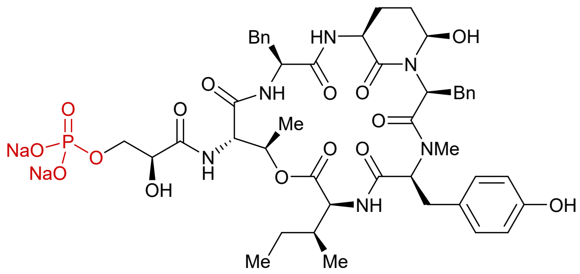


Ulosantoin (**S13**)

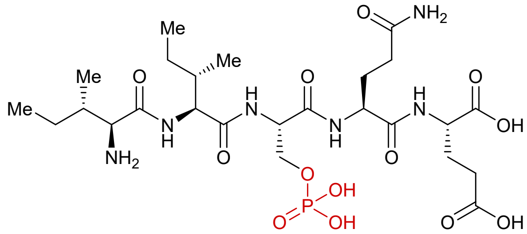


Guanitoxin/anatoxin-a(S) (**S14**)

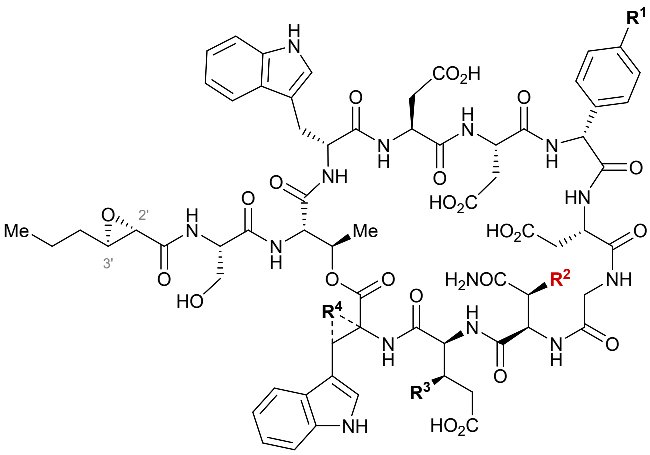
Figure S3. Peptides



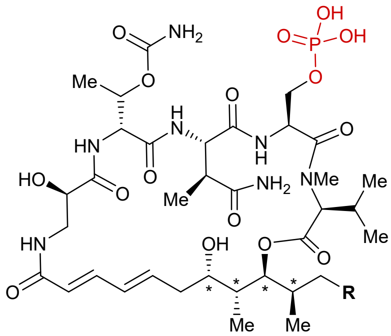
Micropeptin T-20 (**66**)
(Putative structure)



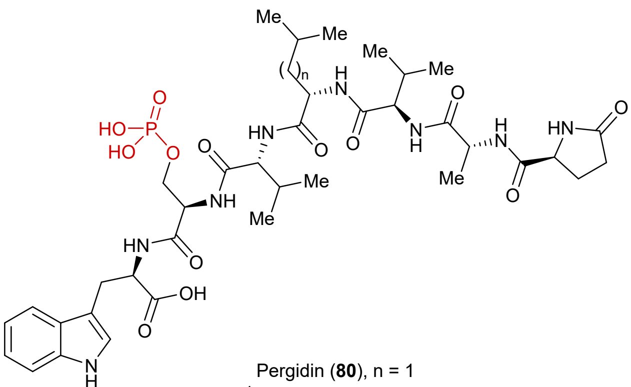
Alphostatin (**S15**)



	R^1	R^2	R^3	R^4
CDA1b (**67**)	OH	OPO_3H_2	H	H,H
CDA2a (**68**)	OH	OPO_3H_2	Me	π-bond
CDA2b (**69**)	OH	OPO_3H_2	Me	H,H
CDA2d (**70**)	H	OPO_3H_2	Me	H,H
CDA2fa (**75**)	F	OPO_3H_2	Me	π-bond
CDA2fb (**76**)	F	OPO_3H_2	Me	H,H



	R
Celebeside A (**77**)	Et
Celebeside B (**78**)	Me

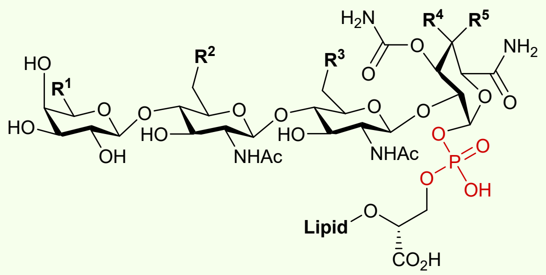


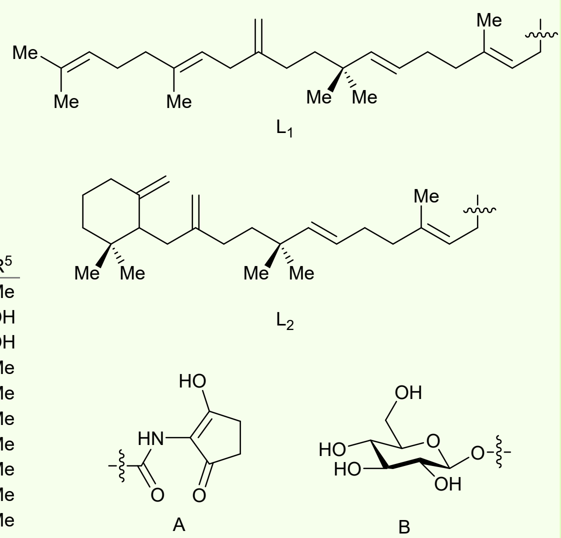
Pergidin (**80**), n = 1
Val[4]-pergidin (**81**), n = 0

*Absolute configuration not determined.

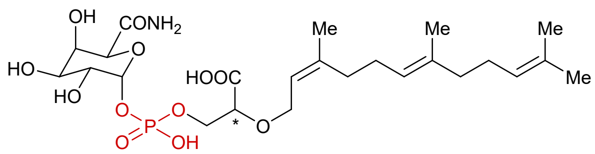
Figure S4. Carbohydrate-based Secondary Metabolites

Moenomycin Family

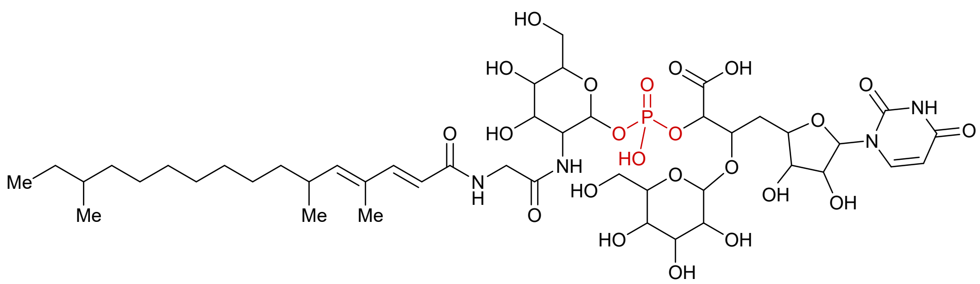




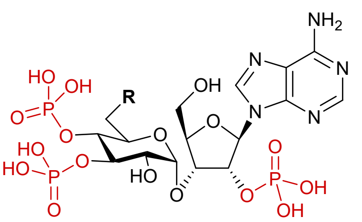
	Lipid	R^1	R^2	R^3	R^4	R^5
Moenomycin A (**82**)	L_1	A	H	B	OH	Me
Moenomycin A_{12} (**S16**)	L_1	A	H	B	H	OH
Moenomycin C_1 (**S17**)	L_1	A	H	H	H	OH
Moenomycin C_3 (**S18**)	L_1	A	H	H	OH	Me
Moenomycin C_4 (**S19**)	L_1	A	H	OH	OH	Me
AC326-α (**83**)	L_2	A	OH	B	OH	Me
Nosokomycin A (**84**)	L_1	COOH	H	B	OH	Me
Nosokomycin B (**85**)	L_1	$CONH_2$	H	B	OH	Me
Nosokomycin C (**86**)	L_1	COOH	H	OH	OH	Me
Nosokomycin D (**87**)	L_1	$CONH_2$	H	OH	OH	Me
Pholipomycin (**S20**)	L_1	A	OH	OH	OH	Me



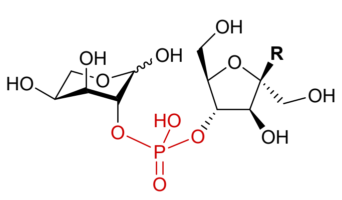
Nosokophic acid (**88**)



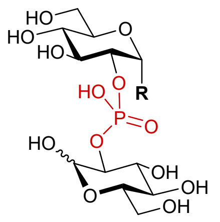
A-94964 (**89**)
(Unknown stereochemistry)



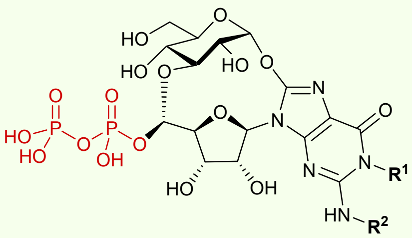
	R
Adenophostin A (**90**)	OH
Adenophostin B (**91**)	OAc



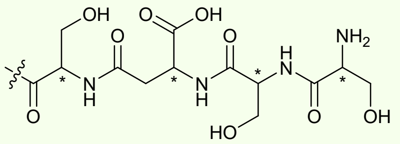
	R
Agrocinopine A (**92**)	α-Glc
Agrocinopine B (**93**)	OH

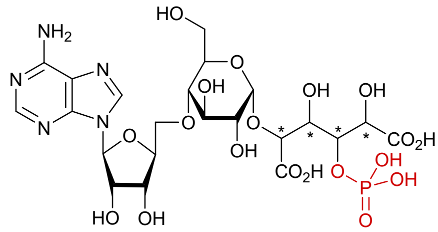


	R
Agrocinopine C (**94**)	β-Fru
Agrocinopine D (**95**)	(α/β)-OH



	R^1	R^2
Guanofosfocin A (**S21**)	Me	H
Guanofosfocin B (**S22**)	H	H
Guanofosfocin C (**S23**)	H	A

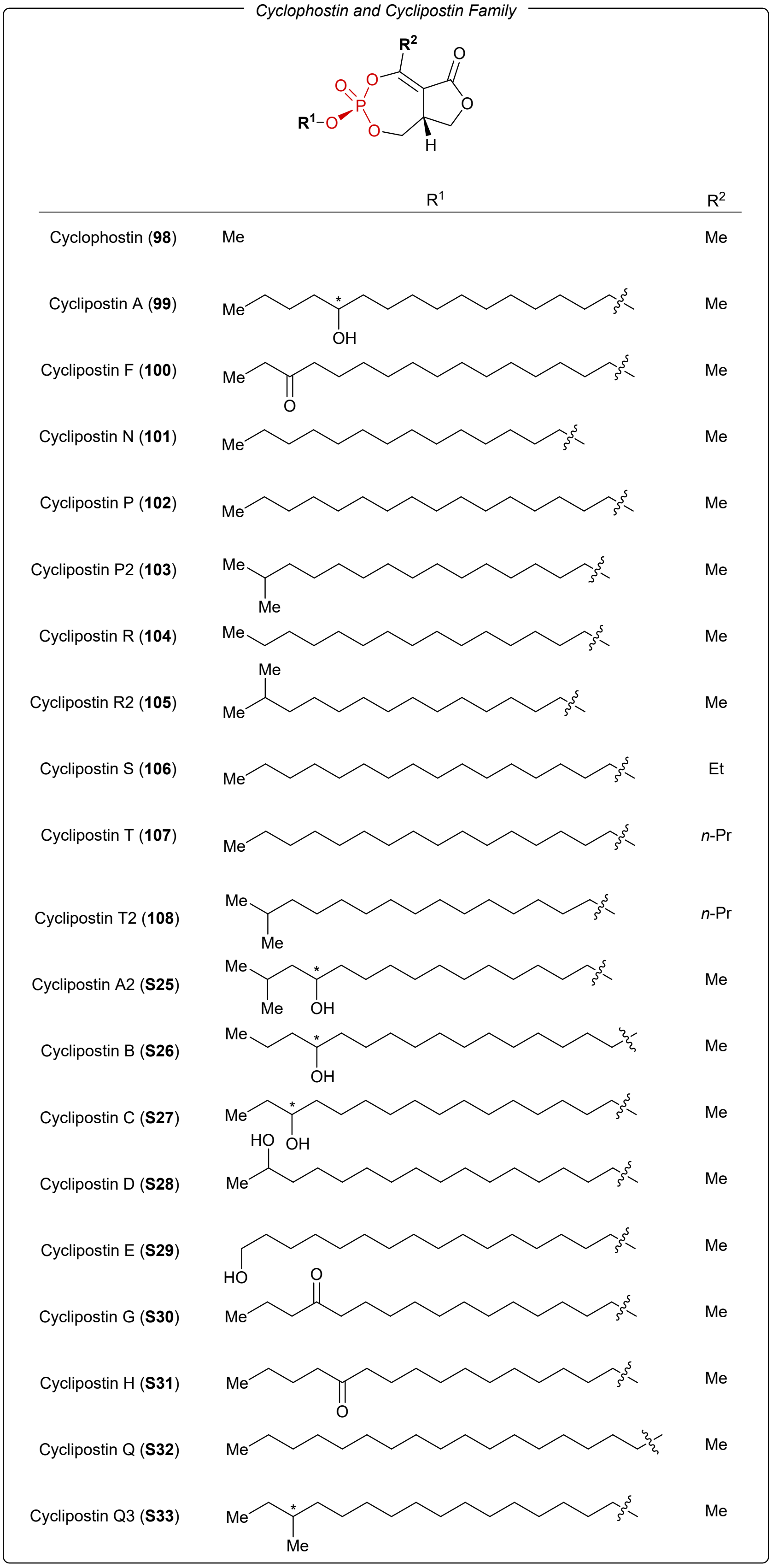
A = 

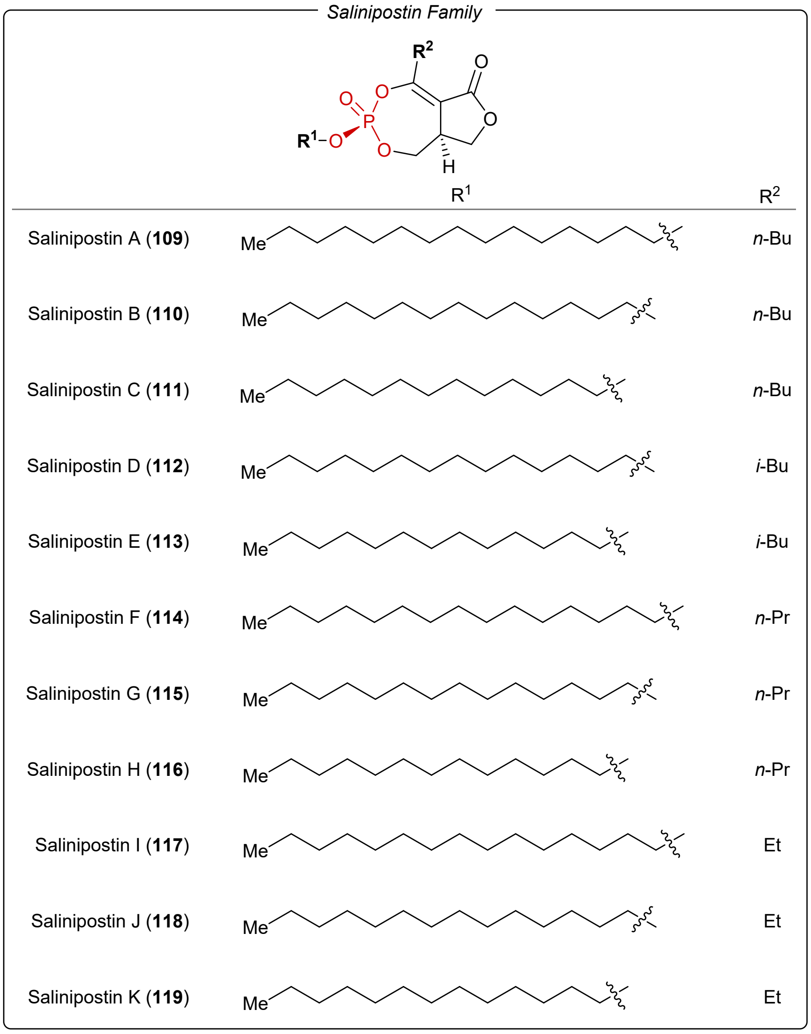


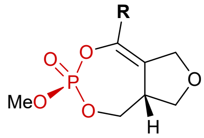
Thuringiensin (**S24**)

*Absolute configuration not determined.

Figure S5: Enolphosphates





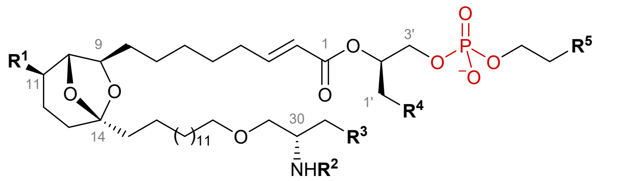


CGA 134,735 (**96**) R = n-C_3H_7
CGA 134,736 (**97**) R = Me

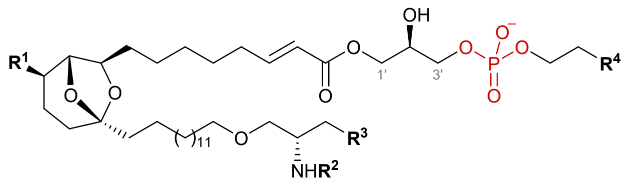
*Absolute configuration not determined.

Figure S6. Lipid-based Secondary Metabolites

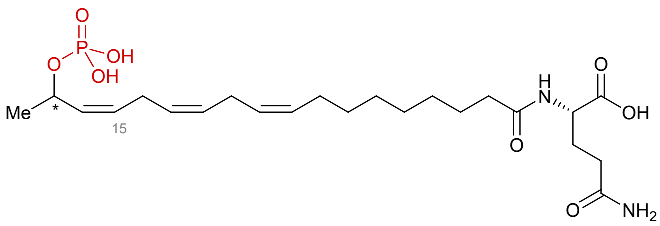
Siladenoserinol Family



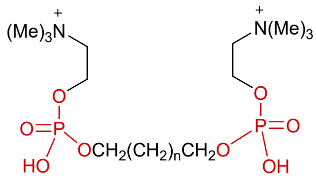
Siladenoserinol	R1	R2	R3	R4	R5
A (**120**)	OAc	SO_3H	OH	OAc	NMe_3^+
B (**121**)	OAc	SO_3H	OH	OAc	NH_3^+
C (**122**)	OH	H	OSO_3H	OAc	NH_3^+
D (**123**)	OAc	SO_3H	OH	OH	NMe_3^+
E (**124**)	OH	H	OSO_3H	OAc	NMe_3^+
F (**125**)	OH	SO_3H	OH	OAc	NMe_3^+
G (**126**)	OAc	H	OSO_3H	OAc	NMe_3^+



Siladenoserinol	R1	R2	R3	R4
H (**127**)	OAc	SO_3H	OH	NMe_3^+
I (**128**)	OAc	H	OSO_3H	NMe_3^+
J (**129**)	OH	H	OSO_3H	NH_3^+
K (**130**)	OH	SO_3H	OH	NMe_3^+
L (**131**)	OH	H	OSO_3H	NMe_3^+



N-(17-Phosphonooxylinolenoyl)-L-glutamine (**132**)
N-(17-phosphonooxylinoleoyl)-L-glutamine (**133**) (saturated at C15-C16)

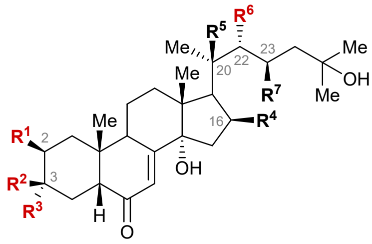


Irlbacholine (**135**) n = 20
Gentianaline A (**136**) n = 19
Gentianaline B (**137**) n = 21
Gentianaline C (**138**) n = 22

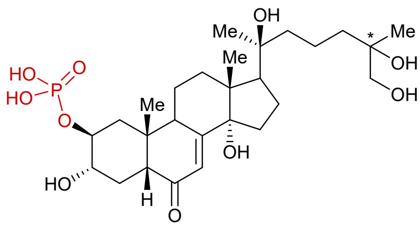
*Absolute configuration not determined.

Figure S7. Steroids

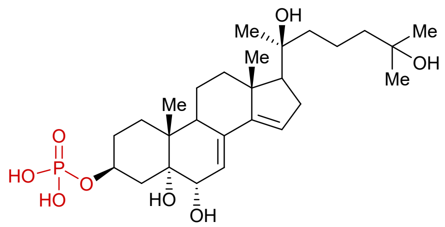
Ecdysone Family



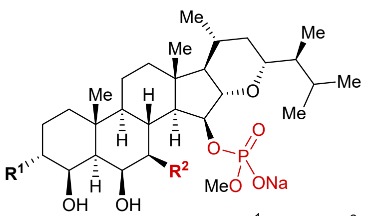
	R^1	R^2	R^3	R^4	R^5	R^6	R^7
Ecdysone 22-phosphate (**S34**)	OH	OH	H	H	H	OPO_3H_2	H
2-Deoxyecdysone 22-phosphate (**S35**)	H	OH	H	H	H	OPO_3H_2	H
20-Hydroxyecdysone 22-phosphate (**S36**)	OH	OH	H	H	OH	OPO_3H_2	H
2-Deoxy-20-hydroxyecdysone 22-phosphate (**S37**)	H	OH	H	H	OH	OPO_3H_2	H
20-Hydroxyecdysone 3-acetate 22-phosphate (**S38**)	OH	OAc	H	H	OH	OPO_3H_2	H
3-Epi-2-deoxyecdysone 3-phosphate (**S39**)	H	H	OPO_3H_2	H	H	OH	H
22-Deoxy-20-hydroxyecdysone 3-phosphate (**S40**)	OH	OPO_3H_2	H	H	OH	H	H
3-Epi-22-deoxy-20-hydroxyecdysone 2-phosphate (**S41**)	OPO_3H_2	H	OH	H	OH	H	H
2,22-Dideoxy-20-hydroxyecdysone 3-phosphate (**S42**)	H	OPO_3H_2	H	H	OH	H	H
3-Epi-22-deoxy-16β,20-dihydroxyecdysone 2-phosphate (**140**)	OPO_3H_2	H	OH	OH	OH	H	H
2,22-dideoxy-23-hydroxyecdysone 3-phosphate (**141**)	H	OPO_3H_2	H	H	H	H	OH



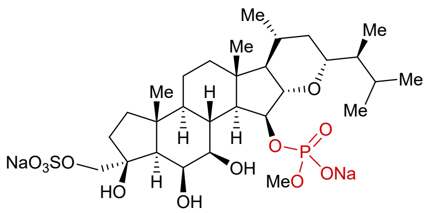
3-Epi-22-deoxy-20,26-dihydroxyecdysone 2-phosphate (**139**)



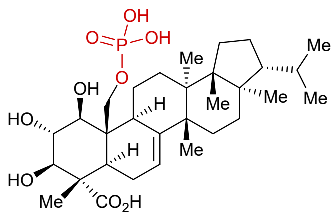
Bombycosterol 3-phosphate (**S43**)



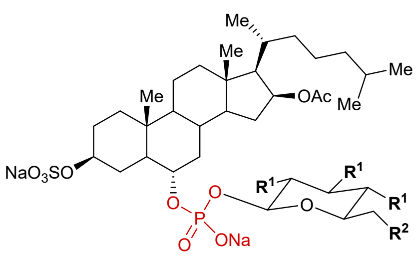
	R^1	R^2
Haplosamate A (**142**)	OSO_3Na	OH
Haplosamate B (**143**)	OSO_3Na	OPO_3Na_2
Desulfohaplosamate (**144**)	OH	OH



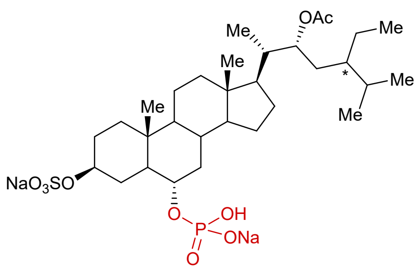
145



RPR113228 (**S44**)



	R^1	R^2
Tremasterol A (**S45**)	OH	OH
Tremasterol B (**S46**)	OAc	OAc
Tremasterol C (**S47**)	OH	OAc



Tremasterol D (**S48**)

*Absolute configuration not determined.

Table S1. Natural sources of phosphorylated secondary metabolites (extended)

Class	Natural Products	Source	Ref.
Polyketides	Fostriecin (**1**) PD 113,270 (**2**) PD 113,271 (**3**)	*Streptomyces pulveraceus* ATCC 31906	1-5
	PD 114,027 (**S1**)	*Streptomyces pulveraceus* ATCC 31906	2, 6, 7
	Phostriecin (aka Sultriecin) (**4**)	*Streptomyces roseiscleroticus* L827-7 ATCC 53903	8, 9
	Phosdiecin A (**5**) Phosdiecin B (**6**)	*Streptomyces* sp. SS99BA-2	10
	S2 (unnamed compound)	*Streptomyces hygroscopicus* MA-5000	11
	Cytostatin (**7**)	*Streptomyces* sp. MJ654-NF4	12, 13
	Phosphazomycin A (structure has not yet been reported)	*Streptomyces* sp. HK-803	14
	Phoslactomycin A (**8**)	*Streptomyces nigrescens* SC-273	15, 16
	Phoslactomycin B (Phospholine) (**9**)	*Streptomyces nigrescens* SC-273; *Streptomyces hygroscopicus*	15-18
	Phoslactomycin C/Phosphazomycin C_1 (**10**)	*Streptomyces* sp. HK-803; *Streptomyces nigrescens* SC-273	15, 16, 19
	Phosphazomycin C_2 (**S3**)	*Streptomyces* sp. HK-803	19
	Phoslactomycin D (**11**)	*Streptomyces nigrescens* SC-273	15, 16
	Phoslactomycin E (**12**)	*Streptomyces nigrescens* SC-273; *Streptomyces* sp. HA81-2	15, 16, 20
	Phoslactomycin F (**13**)	*Streptomyces nigrescens* SC-273; *Streptomyces platensis* SANK 60191	15, 16, 21-23
	Phoslactomycin G/Leustroducsin H (**14**)	*Streptomyces* sp. HK 803	24
	Leustroducsin A (**15**) Leustroducsin B (**16**) Leustroducsin C (**17**)	*Streptomyces platensis* SANK 60191	21-23
	Phoslactomycin H (**18**) Phoslactomycin I (**19**)	*Streptomyces* sp. MLA1839	25
	S4 (unnamed compound)	*Streptomyces platensis* SAM-0654	26
	I-h (**S5**) I-i (**S6**) I-j (**S7**)	*Streptomyces platensis* SANK 60191	21, 22
	S8 (unnamed compound)	*Streptomyces platensis* SAM-0654	26
	Franklinolide A (**20**)	Australian marine sponge CMB-01989; *Bahamian sponge Plakina jamaicensis*	27, 28
	Franklinolide B (**21**) Franklinolide C (**22**)	Australian marine sponge CMB-01989	27
	(–)-Enigmazole B (**S9**)	*Cinachyrella enigmatica*	29
	Difficidin (**S10**)	*Bacillus subtilis* ATCC 39320; *Bacillus subtilis* ATCC 39374; *Bacillus subtilis* A1/3; *Bacillus amyloliquefaciens*; *Bacillus methylotrophicus* DR-08	30-35
	Oxydifficidin (**S11**)	*Bacillus subtilis* ATCC 39320; *Bacillus subtilis* ATCC 39374; *Bacillus subtilis* A1/3; *Bacillus amyloliquefaciens*; *Bacillus methylotrophicus* DR-08	30, 31, 33, 35
	Calyculin A (**23**)	*Discodermia calyx*; *Lamellomorpha strongylata*	36, 37
	Calyculin B (**24**)	*Discodermia calyx*; *Lamellomorpha strongylata*	37, 38
	Calyculin C (**25**)	*Discodermia calyx*	38
	Calyculin D (**26**)	*Discodermia calyx*	38
	Calyculin E (**27**)	*Lamellomorpha strongylata*;	37, 39,

		Discodermia sp.; *Discodermia calyx*	40
	Calyculin F (**28**)	*Lamellomorpha strongylata*; *Discodermia* sp.; *Discodermia calyx*	37, 39, 40
	Calyculin G (**29**)	*Discodermia calyx*	40
	Calyculin H (**30**)	*Discodermia calyx*	40
	Des-*N*-methylcalyculin A (**31**)	*Discodermia calyx*	41
	Calyculinamide A (**32**)	*Lamellomorpha strongylata*; *Discodermia calyx*	37, 41
	Calyculinamide B (**33**)	*Lamellomorpha strongylata*	37
	Calyculinamide F (**34**)	*Discodermia calyx*	41
	Phosphocalyculin A (**36**)	*Discodermia calyx*	42, 43
	Phosphocalyculin C (**37**)	*Discodermia calyx*	42
	Calyculin J (**38**)	*Discodermia calyx*	41
	Hemicalyculin A (**39**)	*Discodermia calyx*	44
	Clavosine A (**40**) Clavosine B (**41**) Clavosine C (**42**)	*Myriastra clavosa*	45
	Geometricin A (**43**)	*Luffariella geometrica*	46
	Swinhoeiamide A (**44**)	*Theonella swinhoei*	47, 48
	Phosphoeleganin (**45**)	*Sidnyum elegans*	49, 50
	Enigmazole A (**46**) 15-*O*-Methylenigmazole A (**47**) 13-Hydroxy-15-*O*-methylenigmazole A (**48**)	*Cinachyrella enigmatica*	51
	Eushearilide (**49**)	*Eupenicillium shearii* IFM54447	52, 53
	JBIR-19 (**50**) JBIR-20 (**51**)	*Metarhizium* sp. fE61	54
	Preussolide A (**52**) Preussolide B (**53**)	*Preussia typharum*	55
Alkaloids and Related Secondary Metabolites	Psilocybin (**54**)	*Psilocybe argentipes;* *Psilocybe baeocystis*; *Psilocybe cubensis*; *Inocybe* (Fr.) Fr.; *Psilocybe semilanceata*; *Psilocybe medullosa*; *Psilocybe mexicana*; *Psilocybe cyanescens*; *Psilocybe bohemica Sebek*; *Psilocybe liniformans*; *Panaeolus subbalteatus*; *Panaeolus cyanescens*; *Inocybe aeruginascens*; *Inocybe corydalina*; *Inocybe coelestium*; *Inocybe haemacta*; *Gymnopilus purpuratus*; *Pluteus salicinus*; *Pholiotina cyanopus*	56-69
	Baeocystin (**57**)	*Psilocybe baeocystis*; *Psilocybe semilanceata*; *Psilocybe cubensis*; *Psilocybe bohemica*; *Inocybe* (Fr.) Fr.; *Pholiotina cyanopus*; *Inocybe aeruginascens*; *Gymnopilus purpuratus*; *Panaeolus cyanescens*	56-62, 70-72
	Norbaeocystin (**58**)	*Psilocybe baeocystis*;	58, 59,

		Psilocybe cubensis; *Pholiotina cyanopus*	71
	Aeruginascin (**59**)	*Inocybe aeruginascens*; *Psilocybe cubensis*; *Pholiotina cyanopus*	58-60, 73
	Amicoumacin A 8'-phosphate (**60**) Amicoumacin B 8'-phosphate (**61**)	*Bacillus pumilus*	74
	FR901483 (**64**)	*Cladobotryum* sp. 11231	75
	Tagetitoxin (**65**)	*Pseudomonas syringae* pv. *tagetis*	76, 77
	Pokepola ester (**S12**)	*Spongia oceania*	78
	Ulosantoin (**S13**)	*Ulosa ruetzleri*	79
	Guanitoxin (aka anatoxin-a(S)) (**S14**)	*Anabaena spiroides*; *Anabaena flos-aquae*; *Anabaena lemmermannii*; *Anabaena crassa*	80-88
Peptides	Micropeptin T-20 (**66**)	*Microcystis aeruginosa*	89, 90
	Alphostatin (**S15**)	*Bacillus megaterium* BMG59-R2	91, 92
	CDA1b (**67**) CDA2a (**68**) CDA2b (**69**)	*Streptomyces coelicolor* A3(2) 2377	93-95
	CDA2d (**70**) CDA2fa (**75**) CDA2fb (**76**)	*Streptomyces coelicolor* A3(2) 2377Δ*hmaS* mutant	94
	Celebeside A (**77**) Celebeside B (**78**)	*Siliquariaspongia mirabilis*	96
	Pergidin (**80**)	*Perreyia flavipes*; *Philomastix macclaei*; *Lophyrotoma analis*; *Lophyrotoma interrupta*; *Lophyrotoma zonalis*; *Pterygophorus insignis*; *Pterygophorus* nr *turneri*; *Arge nigripes*; *Arge pagana*; *Arge aruncus*; *Arge captiva*; *Arge dimidiata*; *Arge enkianthus*; *Arge gracilicornis*; *Arge mali*; *Arge melanochra*; *Arge meliosmae*; *Arge nigripes*; *Arge nigronodosa*; *Arge obesa*; *Arge ochropus*; *Arge pagana*; *Arge pullata*; *Arge rejecta*; *Arge similis*; *Arge suspicax*; *Arge suzukii*; *Arge ustulata*; *Arge zonalis*; *Spinarge affinis*; *Spinarge fulvicornis*; *Spinarge nigricornis*; *Spinarge prunivora;* *Cibdela janthina*; *Schizocerella pilicornis*	97-100

	4-Valinepergidin (**81**)	*Philomastix macclaei*; *Lophyrotoma analis*; *Lophyrotoma interrupta*; *Lophyrotoma zonalis*; *Pterygophorus insignis*; *Pterygophorus* nr *turneri*; *Arge dimidiata*; *Arge enkianthus*; *Arge mali*; *Arge rejecta*; *Arge similis*; *Arge suspicax*; *Arge suzukii*; *Spinarge affinis*; *Spinarge fulvicornis*; *Spinarge nigricornis*; *Spinarge prunivora*	100
Carbohydrate-based Secondary Metabolites	Moenomycin A (**82**)	*Streptomyces ghanaensis* ATCC 14672; *Streptomyces bambergiensis* ATCC 13879; *Streptomyces ederensis* ATCC 15304; *Streptomyces geysiriensis* ATCC 15303; Moenomycin complex (Flavomycin®)	101-110
	Moenomycin A_{12} (**S16**)	*Streptomyces ghanaensis* ATCC 14672; Moenomycin complex (Flavomycin®)	103, 104, 109, 111
	Moenomycin C_1 (**S17**)	*Streptomyces ghanaensis* ATCC 14672; Moenomycin complex (Flavomycin®)	103, 104, 109, 112
	Moenomycin C_3 (**S18**)	*Streptomyces ghanaensis* ATCC 14672; Moenomycin complex (Flavomycin®)	103, 104, 109, 113
	Moenomycin C_4 (**S19**)	*Streptomyces ghanaensis* ATCC 14672; Moenomycin complex (Flavomycin®)	103, 104, 109, 113
	AC326-α (**83**)	unidentified *Actinomyces* sp. AC326	103, 114
	Nosokomycin A (**84**) Nosokomycin B (**85**)	Moenomycin complex (Flavomycin®); *Streptomyces* sp. K04-0144	103, 109, 115, 116
	Nosokomycin C (**86**) Nosokomycin D (**87**)	*Streptomyces* sp. K04-0144	103, 115, 116
	Pholipomycin (**S20**)	*Streptomyces lividoclavatus*	103, 113, 117-121
	Nosokophic acid (**88**)	*Streptomyces* sp. K04-0144	122
	A-94964 (**89**)	*Streptomyces* sp. SANK 60404	123, 124
	Adenophostin A (**90**) Adenophostin B (**91**)	*Penicillium brevicompactum* SANK 11991; *Penicillium brevicompactum* SANK 12177	125-127
	Agrocinopine A (**92**)	tumours induced by nopaline strains	128-130
	Agrocinopine B (**93**)	tumours induced by nopaline strains	128
	Agrocinopine C (**94**)	tumours induced by agropine strains	128, 131
	Agrocinopine D (**95**)	tumours induced by agropine strains	128
	Guanofosfocin A (**S21**)	*Streptomyces sp.* AB2570	132
	Guanofosfocin B (**S22**)	*Trichoderma sp.* FD5372	132
	Guanofosfocin C (**S23**)	*Trichoderma sp.* FD5372	132
	Thuringiensin (aka β-exotoxin) (**S24**)	*Bacillus thuringiensis*	133-138
Enolphosphates	CGA 134,735 (**96**)	*Streptomyces antibioticus* DSM 1951	139

	CGA 134,736 (**97**)		
	Cyclophostin (**98**)	*Streptomyces lavendulae* NK901093	140
	Cyclipostin A (**99**) Cyclipostin F (**100**) Cyclipostin N (**101**) Cyclipostin P (**102**) Cyclipostin P2 (**103**) Cyclipostin R (**104**) Cyclipostin R2 (**105**) Cyclipostin S (**106**) Cyclipostin T (**107**) Cyclipostin T2 (**108**) Cyclipostin A2 (**S25**)* Cyclipostin B (**S26**)* Cyclipostin C (**S27**)* Cyclipostin D (**S28**)* Cyclipostin E (**S29**)* Cyclipostin G (**S30**)* Cyclipostin H (**S31**)* Cyclipostin Q (**S32**)* Cyclipostin Q3 (**S33**)* *HPLC detected	*Streptomyces* sp. DSM 13381	141-143
	Salinipostin A (**109**) Salinipostin B (**110**) Salinipostin C (**111**) Salinipostin D (**112**) Salinipostin E (**113**) Salinipostin F (**114**) Salinipostin G(**115**) Salinipostin H (**116**) Salinipostin **I** (**117**) Salinipostin J (**118**) Salinipostin K (**119**)	*Salinispora* sp. RL08-036-SPS-B	144
Lipid-based Secondary Metabolites	Siladenoserinol A (**120**) Siladenoserinol B (**121**) Siladenoserinol C (**122**) Siladenoserinol D (**123**) Siladenoserinol E (**124**) Siladenoserinol F (**125**) Siladenoserinol G (**126**) Siladenoserinol H (**127**) Siladenoserinol **I** (**128**) Siladenoserinol J (**129**) Siladenoserinol K (**130**) Siladenoserinol L (**131**)	Didemnidae tunicate	145
	N-(17-Phosphonooxylinolenoyl)glutamine (**132**) *N*-(17-phosphonooxylinoleoyl)glutamine (**133**)	*Spodoptera exigua*	146
	Irlbacholine (**135**)	*Irlbachia alata*; *Anthocleista djalonensis*; *Gentiana crassicaulis*; *Gentiana macrophylla*; *Gentiana dahurica*; *Gentiana straminea*	147, 148
	Gentianaline A (**136**) Gentianaline B (**137**) Gentianaline C (**138**)	*Gentiana crassicaulis*; *Gentiana macrophylla*; *Gentiana dahurica*; *Gentiana straminea*	148
Steroids	Ecdysone 22-phosphate (**S34**)	*Schistocerca gregaria*; *Bombyx mori*	149-151

	2-Deoxyecdysone 22-phosphate (**S35**)	*Schistocerca gregaria*; *Locusta migratoria*; *Ascaris lumbricoides*; *Bombyx mori*	149-153
	20-Hydroxyecdysone 22-phosphate (**S36**)	*Schistocerca gregaria*; *Locusta migratoria*; *Bombyx mori*	149, 151, 154
	2-Deoxy-20-hydroxyecdysone 22-phosphate (**S37**)	*Schistocerca gregaria*; *Locusta migratoria*; *Bombyx mori*	149, 151, 154
	20-Hydroxyecdysone 3-acetate 22-phosphate (**S38**)	*Locusta migratoria*	154
	3-Epi-2-deoxyecdysone 3-phosphate (**S39**)	*Locusta migratoria*	152, 155
	22-Deoxy-20-hydroxyecdysone 3-phosphate (**S40**)	*Bombyx mori*	156
	3-Epi-22-deoxy-20-hydroxyecdysone 2-phosphate (**S41**)	*Bombyx mori*	157
	2,22-Dideoxy-20-hydroxyecdysone 3-phosphate (**S42**) Bombycosterol 3-phosphate (**S43**)	*Bombyx mori*	151, 158
	3-Epi-22-deoxy-20,26-dihydroxyecdysone 2-phosphate (**139**) 3-Epi-22-deoxy-16β,20-dihydroxyecdysone 2-phosphate (**140**)	*Bombyx mori*	159
	2,22-dideoxy-23-hydroxyecdysone 3-phosphate (**141**)	*Bombyx mori*	160
	Haplosamate A (**142**)	*Xestospongia* sp.; *Cribrochalina* sp.; *Dasychalina* sp.	161-163
	Haplosamate B (**143**)	*Xestospongia* sp.; *Cribrochalina* sp.	161, 162
	Desulfohaplosamate (**144**)	*Dasychalina* sp.	163
	145 (unnamed compound)	*Cribrochalina* sp.	162
	RPR113228 (**S44**)	*Chrysosporium lobatum*; *Chrysosporium* sp.	164, 165
	Tremasterol A (**S45**) Tremasterol B (**S46**) Tremasterol C (**S47**) Tremasterol D (**S48**)	*Tremaster novaecaledoniae*	166, 167

Table S2. Calyculin A (23): Biological Studies

	Ref.
Inhibited NK cell function in mice	168
Inhibited cytoplasmic streaming and induced morphological changes in the cytoplasm of root hair cells of *L. stoloniferum*	169
Induced focal adhesion assembly and tyrosine phosphorylation of non-receptor tyrosine kinase $p125^{Fak}$, of adapter proteins $p130^{Cas}$, and paxillin in Swiss 3T3 cells	170
Myosin activity is necessary for calyculin A-induced cytoplasmic spheric bodies formation in root hair cells of *L. stoloniferum*	171
Calyculin A (**23**) had multiple effects on chromosome movements during anaphase and altered the distribution of spindle proteins in crane-fly spermatocytes	172
Induced actin phosphorylation and depolymerization in renal epithelial cells	173
Promoted activation of several endogenous platelet protein kinases	174
Promoted retraction of mature megakaryocytes proplatelets derived from murine embryonic stem cells	175
Modulated activation markers in TRAP-stimulated human platelets	176
Impaired clot retraction, platelet activation and thrombin generation in human platelets	177
Induced inhibition of the transition from mitosis to interphase in *C. japonicus* eggs	178
Promoted activation of the signalling pathways involving mitogen-activated protein kinase $p38^{MAPK}$ and mitochondrial Ca^{2+}-mediated oxidative stress	179
Induced inhibition of PP2A, activation of glycogen synthase kinase-3 (GSK-3) in wild type neuroblastoma N2a cells, and tau hyperphosphorylation at both PHF-1 and tau-1 epitopes	180
Hyperphosphorylation of tau protein and neurofilaments induced by calyculin A (**23**) in neuroblastoma N2a cells was reversed by the alkaloid berberine	181
Stimulated the expression of TNF-α mRNA in mouse osteoblastic MC3T3-E1 cells	182
Increased the expression of TNF-α mRNA and activation of NF-κB *via* phosphorylation and nuclear translocation of NF-κB in primary osteoblasts from new-born rat calvaria	183
Combination of calyculin A (**23**) and celecoxib inhibited epithelial-mesenchymal transition in human oral squamous cell carcinoma (HSC-3 cells)	184
Promoted phosphorylation of Ser473 in protein kinase B/AkT supporting levels of AkT activation like those found under physiological serum-growth conditions	185
Inhibited expression of CD8α but not of CD4 in human peripheral blood T cells	186
Blocked ERK5 activation by the epidermal growth factors and oxidative stress	187
Suppressed the decline in protein phosphorylation state and increased an inactive form of PP1 in the post-acrosomal region of boar spermatozoa	188
Calyculin A-sensitive protein phosphatases (PP1/PP2A) suppressed full activation of protein kinase A, and enhanced the phosphorylation state of flagellar proteins in sperm flagella of mouse epididymal spermatozoa	189
Induced association of vimentin with 14-3-3 protein family in COS-7 cells	190
Induced centrosome-directed translocation of Weibel-Palade bodies in human aortic endothelial cells	191
Induced elongation of microvilli in unfertilized sea urchin eggs	192
Induced cortical contraction in unfertilized sea urchin eggs	193
Promoted reorganization of the actin cytoskeleton and the cell centre in sea urchin coelomocytes	194
Promoted activation of histone H_1 kinase and condensation of chromosomes in unfertilized sea urchin eggs	195
Melatonin protected SH-SY5Y neuroblastoma cells from calyculin A-induced neurofilament impairment and neurotoxicity	196
Injection of calyculin A (**23**) into rat hippocampus bilaterally led to lesions in spatial memory retention	197

Induced neurite retraction in cultured hippocampal neurons	198
Intraperitoneal administration of melatonin before injection of calyculin A (**23**) prevented calyculin A-induced synaptophysin loss, memory retention deficits and hyperphosphorylation of tau protein and neurofilaments	199
Suppressed *in vivo* early elimination of histone γ-H2AX and DNA double-strand breaks repair	200
Inhibited γ-H2AX dephosphorylation involved in double-strand breaks	201
Calyculin A (**23**) was used as a useful tool to mark the induced γ-H2AX foci after low dose irradiation of lymphocytes	202
Enhanced cyclic D1 phosphorylation and degradation, and arrested cell cycle progression in human breast cancer cells	203
Calyculin A (**23**) acted as a dual action toxin that blocked calcium influx and inhibited protein Ser/Thr phosphatases	204
Combination of calyculin A (**23**) and zoledronic acid showed synergistic cytotoxicity and apoptosis against MCF-7 and MDA-MB-231 breast cancer cells	205
Inhibited potassium channel currents in vascular smooth muscle cells by a mechanism not mediated by the inhibition of protein phosphatases	206
Calyculin A (**23**) doubled the twitch magnitude and increased its duration by 50% in field-stimulated cells but did not significantly increase sarcoplasmic reticulum Ca^{2+} uptake	207
Disrupted sub-plasmalemmal junction and recurring Ca^{2+} waves in vascular smooth muscle	208
Calyculin A-sensitive phosphatases dephosphorylated the sodium-hydrogen exchanger 3 protein (NHE3) altering its activity in opossum kidney cells	209
Enhanced apoptosis mediated by tumour necrosis factor-related apoptosis-inducing ligand (TRAIL) in human renal carcinoma cell line (Caki)	210
Triggered cell shrinkage and phospholipid scrambling of the erythrocyte cell membrane	211
Inhibited nitrergic relaxations of the mouse anococcygeus muscle	212
Decreased Ca^{2+} concentration in rat basophilic leukemic cells during activation of the Ca^{2+} release–activated Ca^{2+} entry pathway	213
Induced endothelial cell shape changes in bovine pulmonary artery endothelial cells	214
Calyculin A (**23**) revealed the role of protein serine/threonine phosphatases in controlling the transcriptional activity of STAT 3	215
Prevented DNA fragmentation, Bax translocation, caspase 8 cleavage, and Bid degradation in human myeloma leukemic cells (ML-1 cells)	216
Induced expression of PTEN, a tumour suppressor gene encoding a phosphatase, through activation of EGR-1 during apoptosis in human squamous carcinoma cells	217
Induced apoptosis in human osteoblastic MG63 cells, elicited the phosphorylation of NF-kB on Ser-536 in human osteoblastic MG63 cells, promoting transcriptional activity of NF-kB related genes	218
NADPH oxidase was not involved in the calyculin A-mediated inhibition of Ca^{2+} influx into neutrophils	219
Treatment of pollen tubes of *L. longiflorum* with calyculin A (**23**) after germination arrested growth, reversibly altered the alignment of actin bundles, and disassembled microtubules	220
Induced subapical membrane aggregation and actin baskets supporting retrograde membrane flow in *L. longiflorum*	221
A calyculin A-sensitive protein phosphatase is required for *B. anthracis* lethal toxin induced cytotoxicity	222
The phosphorylation level of prohibitin, a chaperone involved in the assembly of mitochondrial respiratory chain complex, was increased after calyculin A (**23**) treatment in a rice lesion-mimic mutant in *cdr1*	223

Table S3. Adenophostin A (90): Biological Studies

	Ref.
Rescued inhibition of myristoylated alanine-rich C kinase substrate in permeabilized human sperm	224
Activated plasma membrane large-conductance Ca^{2+}- activated K^{+} channels in mice cerebral artery smooth muscle cells	225
Application of AdA (**90**) into stratum pyramidale non-pyramidal neurons in rat hippocampal slices enhanced excitatory post-synaptic currents on non-pyramidal neurons in the stratum pyramidale and occluded paired stimuli-induced synaptic potentiation	226
Post-synaptic infusions of AdA (**90**) enhanced type A GABA receptor-gated inhibitory postsynaptic currents	227
Increased peak sodium current amplitude in cultured rat striatal neurons	228
Aged mouse eggs injected with AdA (**90**) induced Ca^{2+}-oscillations, caused fragmentation, and other signs of programmed cell death	229
Post-synaptic perfusion of AdA (**90**) increased the probability of detecting synaptic responses during subthreshold presynaptic stimulation and increased spontaneous excitatory postsynaptic current frequency	230
Activation of postsynaptic inositol $InsP_3$ receptors with AdA (**90**) produced a large increase in AMPA receptor excitatory postsynaptic current amplitudes at hippocampal CA1 synapses	231
No oscillations were induced in the $[Ca^{2+}]_i$ at interphase in mouse zygotes	232
Induced temporal degradation of type I $InsP_3$ receptor in bovine oocytes	233
Activated store-operated Ca^{2+} current in rat basophilic leukaemia cells in weak buffer	234
Rescued calcium-induced exocytosis impaired by inhibiting PKC, PLD or sequestering PIP_2, but did not trigger exocytosis	235
Eggs injected with AdA (**90**) showed enhanced Ca^{2+} responses to $CaCl_2$ injections	236
Induced depletion of Ca^{2+} stores in permeabilized microvillar photoreceptors of *H. medicinalis*	237
Treatment of taste receptor cells of the flesh fly *B. peregrina* with AdA (**90**) and deoxycholate elicited the response of the sugar receptor cell	238
Microinjections of AdA (**90**) and $InsP_3$ transiently inhibited growth and induced subapical branches in *N. crassa*	239
Failed to affect bupivacaine anaesthesia in the tail-flick test in mice	240
Injections of AdA (**90**) in nerve terminals of the opener muscle of the first walking leg of *P. clarkii* greatly enhanced synaptic transmission	241
AdA (**90**) was able to mobilize Ca^{2+} from aorta microsomes	242
Activation of $InsP_3$ receptors with AdA (**90**) in permeabilized cells induced Ca^{2+} release in the nuclear domain and other regions of cardiac myocytes isolated from neonatal mouse	243
Increased basal $[Ca^{2+}]_i$ and the frequency of spontaneous Ca^{2+} sparks in saponin-permeabilized myocytes	244
Caused an elevation of 1,4,5-inositol triphosphate-dependent nuclear Ca^{2+} concentration	245
Caused depletion of loaded intracellular Ca^{2+} stores in crypt and surface cells in saponin-permeabilized rat colonic crypts	246
Stimulation of $InsP_3$ receptors *via* intracellular dialysis of AdA (**90**) largely abolished the arachidonic acid-mediated Ca^{2+} signal in rat pancreatic β cells	247
Induced an outburst of ryanodine receptor-dependent spontaneous Ca^{2+} sparks in permeabilized atrial myocytes of atrial fibrillation patients	248
An increase of cytosolic Ca^{2+} concentration within cells was observed when AdA (**90**) encapsulated with 100-nm gold-coated liposomes was injected in the cytosol of single ovarian carcinoma cells	249
Activation of $InsP_3$ receptor by AdA (**90**) was shown to be essential for the opening of mouse transient receptor potential type 5 (mTRP5) in *Xenopus* oocytes.	250

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