

Table S1:

Identification of lead sequences for the molecular evolution of G_{M1}-binding L-peptides. 64 Known or alleged G_{M1} binding motifs obtained from the literature (compiled in [1]) were synthesized as L-peptides on filter membranes and screened for G_{M1} binding with lysoG_{M1}-DY650-conjugate. Fitness values reflect the fluorescence intensities of individual peptide areas as determined in a Licor Odyssey Infrared Imager (arbitrary units). Peptides which showed fluorescence signals significantly above background [2] were selected as lead peptides (written in boldface).

No.	Sequence	Fitness	No.	Sequence	Fitness
1	VEVPGQHIDSQKKAIE	5,14	33	YSEIAIQMMMHMQPKE	7,62
2	VPQGHIDSQKKAIERM	22,09	34	CEFLVKEVTKLIDNNK	50,57
3	GQHIDSQKKAIERMKN	38,86	35	FLVKEVTKLIDNNKTE	57,67
4	HIDSQKKAIERMKNTL	40,54	36	VKEVTKLIDNNKTEKE	17,05
5	DSQKKAIERMKNTLRI	57,76	37	EVTKLIDNNKTEKEIL	5,61
6	SQKKAIERMKNTLRIA	55,99	38	ASQKRPSQRHGSKYLA	56,15
7	SQKKAIERMKNTLRIA	13,64	39	QKRPSQRHGSKYLATA	45,20
8	VPQSQHIDSQKKAIER	24,05	40	RPSQRHGSKYLATAST	36,13
9	GSQHIDSQKKAIERMK	41,53	41	SQRHGSKYLATASTMD	17,07
10	QHIDSQKKAIERMKDT	16,75	42	RHGSKYLATASTMDHA	31,38
11	IDSQKKAIERMKDTLR	33,05	43	GSKYLATASTMDHARH	55,51
12	SQKKAIERMKDTLRIA	37,23	44	KYLATASTMDHARHGF	59,64
13	SYTESMAGKREMVIIIT	9,60	45	LATASTMDHARHGFLP	50,61
14	TESMAGKREMVIIITFK	49,04	46	TASTMDHARHGFLPRH	58,55
15	SMAGKREMVIIITFKSG	48,01	47	STMDHARHGFLPRHRD	49,57
16	LEVPTDGNAGLLAEPQ	5,36	48	MDHARHGFLPRHRDTG	61,29
17	VPTDGNAGLLAEPQIA	6,87	49	HARHGFLPRHRDTGIL	52,37
18	TDGNAGLLAEPQIAMF	8,39	50	RHGFLPRHRDTGILDS	29,82
19	GNAGLLAEPQIAMFCG	17,33	51	GFLPRHRDTGILDSIG	23,39
20	AGLLAEPQIAMFCGRL	30,50	52	LPRHRDTGILDSIGRF	104,05
21	LLAEPQIAMFCGRLNM	27,92	53	KYEVNWKTHEIKVKGQ	63,41
22	AEPQIAMFCGRLNMHM	75,37	54	YEVNWKTHEIKVKGQN	77,44
23	PQIAMFCGRLNMHMNV	83,79	55	AEYHNTQIHTLNDKIF	18,80
24	IAMFCGRLNMHMNVQN	60,97	56	ATFQVEVPGSQHIDSQ	11,26
25	MFCGRLNMHMNVQNGK	56,71	57	AKVEKLCVWNNKTPHA	67,96
26	CGRLNMHMNVQNGKWD	22,38	58	GALSETYSFIKEYHNT	78,21
27	RLNMHMNVQNGKWDS	9,66	59	ESLAGKREQSDIHQSG	12,39
28	NMHMNVQNGKWDS DPS	4,79	60	ASDWYDEMLTWNHGA	66,96
29	DFRRLPGAFWQLRQPA	94,08	61	DGTVSTHECRLEYKYP	43,00
30	GWWYKGRARPVSAAVAA	72,12	62	FSHSGEGDMIWERMID	12,94
31	VWRL LAPPSNRLLPA	64,39	63	HHCSILKEVWHVKKLG	74,12
32	SQYSEIAIQMMMHMQP	9,37	64	PDQDGVVTAMHGFTHN	14,38

Identification of lead sequences for the molecular evolution of G_{M1}-binding D-peptides.

For the evolution of D-peptides the 64 alleged G_{M1} binding motifs of above were transformed from L-peptides into their retro-inverso form (reversal of sequence and use of D-amino acids). Screening of the D-peptides and assignation of fitness values was performed as for the L-peptides.

No.	Sequence	Fitness	No.	Sequence	Fitness
1	eiakkqsdi hqgpvev	1,53	33	ekpqmhmnmqiaiesy	1,84
2	mreiakkqsdi hqgpv	4,35	34	knndilktvekvlfec	5,12
3	nkmreiakkqsdi hqg	10,05	35	etknndilktvekvlf	7,20
4	ltnkmreiakkqsdi h	11,28	36	eketknndilktvekv	0,65
5	irltnkmreiakkqsd	10,09	37	lieketknndilktve	0,20
6	airltnkmreiakkqs	12,97	38	alyksghrqsprkqsa	17,01
7	iakkqsdi hqsgpvev	2,59	39	atalyksghrqsprkq	18,53
8	reiakkqsdi hqsgpv	4,91	40	tsatalyksghrqspr	16,25
9	kmreiakkqsdi hqsg	11,98	41	dmstsatalyksghrqs	11,63
10	tdkmreiakkqsdi hq	6,83	42	ahdmstsatalyksghr	10,07
11	rltdkmreiakkqsd i	5,56	43	hrahdmstsatalyks g	3,90
12	airltdkmreiakkqs	11,73	44	fghrahdmstsataly k	35,71
13	tiivmerkgamsetys	1,63	45	plfghrahdmstsatal	87,02
14	kfttiivmerkgamset	26,79	46	hrplfghrahdmstsat	64,35
15	gskfttiivmerkgams	21,83	47	drhrplfghrahdmsts	90,77
16	qpeallgangdtpvel	1,45	48	gtdrhrplfghrahdm	82,55
17	aiqpeallgangdtpv	1,50	49	ligtdrhrplfghrah	77,87
18	fmai qpeallgangdt	17,73	50	sdligtdrhrplfghr	15,55
19	gc fmai qpeallgang	19,47	51	gisdligtdrhrplfg	15,69
20	lrgcfmai qpeallga	50,05	52	frgisdligtdrhrpl	59,27
21	mn lrgcfmai qpeall	44,35	53	qgkvkiehtkwnveyk	22,36
22	mhmn lrgcfmai qpea	53,31	54	nqgkvkiehtkwnvey	24,24
23	vnmhmn lrgcfmai qp	51,71	55	fikdn lthiqtnhyea	75,51
24	nqvnmhmn lrgcfmai	7,94	56	qsdi hqsgpvevqfta	0,92
25	kg nqvnmhmn lrgcfm	8,74	57	ahptknnwvclkevka	8,22
26	dwkg nqvnmhmn lrgc	4,67	58	tnhyekifsyteslag	102,36
27	dsdwkg nqvnmhmn l r	4,46	59	gsqhidsqerkgalse	-1,23
28	spdsdwkg nqvnmhmn	-1,82	60	aghinwtlmedywdsa	9,73
29	apqrlqwfagplrrfd	157,50	61	pykyelrcehtsvtgd	6,17
30	aavasvprargkywwg	28,12	62	dimrewimdgegshsf	2,29
31	apllrnsfppallrwv	52,06	63	glkkvhwveklischh	12,78
32	pqmhmnmqiaiesyqs	1,57	64	nhtfghmatvvgdqdp	9,23

References:

- [1] Röckendorf N, Bade S, Hirst T R, Gorris H-H, Frey A (2007) Synthesis of a fluorescent ganglioside G_{M1} derivative and screening of a synthetic peptide library for G_{M1} binding sequence motifs. Bioconjugate Chem 18: 573-578.
- [2] Frey A, Di Canzio J, Zurakowski D (1998) A statistically defined endpoint titer determination method for immunoassays. J Immunol Methods 221: 35-41.